Investigations into liquid crystal based optofluidic polarization devices

Ranjini Radhakrishnan

2013

Ranjini Radhakrishnan. (2013). Investigations into liquid crystal based optofluidic polarization devices. Doctoral thesis, Nanyang Technological University, Singapore.

<https://hdl.handle.net/10356/56097>

<https://doi.org/10.32657/10356/56097>

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A thesis submitted to the Nanyang Technological University in partial
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Abstract

Optofluidics, refers to the special type of optical devices fabricated by integrating optics and microfluidics with the aim of manipulating fluid-light interactions. This has developed into one of the latest research areas in the recent past which allows the integration of multiple tasks in a single chip leading to lab-on-a-chip devices. One of the major challenges in developing optofluidic platforms or components is the correct choice of fluid. Though a variety of fluids are used depending upon the applications, the possibilities of incorporating an anisotropic fluid were not well explored. It is to be mentioned that such fluids generally offer additional degrees of freedom and tunability of intended parameters. Based on the analysis of various tunable optofluidic systems reported in literature, it is found that the devices with tunability of optical polarization are a poorly explored area. Further, the inclusion of an anisotropic fluid, liquid crystal, offers a novel method of refractive index modulation. In this context, the main objectives of the thesis focus into the (i) research and development of tunable optofluidic polarization devices, with a novel method of attaining tunability by integrating liquid crystalline materials, (ii) illustration of dynamical properties of liquid crystalline materials and possible effects due to the interaction with light.

Tunability in the state of polarization, based on the flow induced refractive index modulation in the fluid medium, is comparatively a new method which can

offer various possibilities in lab-on-a chip devices fabrication. Also liquid crystals, an efficient choice of fluid for the same are not well explored based on an optofluidic platform. Even though liquid crystals exist in the fluid state at room temperature, an applied shear force or flow can induce an orientational as well as translational motion of the molecules. Thus, due to the molecular realignment there can be refractive index modulation (birefringence) and hence tune the state of polarization of the light beam transmitted through the medium. In order to validate these concepts, both lyotropic and thermotropic liquid crystals are analysed. For lyotropic liquid crystals, hydrated crystals formed are monitored and found to be birefringent which can be controlled with the applied flow. Also the possible fluid-light interactions that can contribute to the transmitted light intensity are investigated.

Thermotropic liquid crystals, in the aligned and non-aligned state are analysed based on the same flow driven perturbations. Scattering losses, apart from polarization is found to be significant for non-aligned liquid crystals. Therefore, to make the liquid crystal medium homogeneous and thereby reducing the scattering effects, a preferential alignment is applied to the liquid crystal molecules. The molecular reorientation occurring across the aligned liquid crystal medium is analysed using conoscopic imaging. The corresponding change in the state of polarization is monitored using optical transmission. Jones matrix of the liquid crystal medium is also formulated based on the alignment of molecules and thereby validating the change in the state of polarization of the transmitted beam.

The proposed concepts and optical configurations for tuning SOP by flow induced molecular reconstruction in liquid crystal can be improved for specific applications in future. Future research will hence be targeted mainly to enhance the device performance and also to explore different device configurations such as variable attenuator, tunable phase retarder and tunable filter. Spectroscopy, imaging and biosensors are the possible application areas in which the flow induced polarization device can be integrated into lab-on-a-chip devices. It is envisaged that the outcome of this thesis can enable future research and development of relevant tunable devices for their potential lab-on-a-chip devices applications.

Acknowledgement

I take this opportunity to acknowledge the help and encouragement received from various sources during the period of my Ph. D programme.

I would like to express my heartfelt thanks to my research supervisor Assoc. Prof. V. M. Murukeshan, who gave the opportunity to pursue Ph.D at a prestigious university. With deep sense of gratitude I express my sincere thanks to him for his excellent guidance, positive criticism and valuable suggestions. I extend my gratitude profoundly to my co-supervisor Assoc. Prof. Nguyen Nam-Trung, for his constant encouragement and creative discussions. I am indebted to both of them for their exemplary guidance and support throughout my research.

I thank Dr. Amrutha and Dr. Shinoj for their support in my experiments. I also express my heartfelt thanks to all my colleagues, staff of Robotic Research Centre for their help, co-operation and support. I acknowledge the research student support received through Nanyang Technological University, Singapore. Also I am thankful for the funding support received through MOE (ARC3/08) for the completion of my Ph.D.

I would like to thank Dr. Rajan K John, lecturer during my Masters, for motivating me to pursue a research career. I am thankful to him for all the achievements in my career.

As a personal note, I would like to thank my husband, Dr. P. Manoj who supported me throughout the Ph.D period. His encouragement and motivation helped to nurture my research career. I thank my parents for their support and help

throughout my life. I wouldn't have been the person who I am today, without their love, blessings and prayers. I also thank my sister for her support and prayers throughout my Ph.D studentship. Finally I am thankful to my son, who really suffered a lot during my studies. Unknowingly, he gave me all support and warmth with his presence. Above all, I thank God almighty for guiding and strengthening me these years.

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List of Symbols

Symbols	Description
δ	Phase difference
Δn	Birefringence
n	Director
m, l	Secondary directors
n_e	Extraordinary refractive index
n_o	Ordinary refractive index
n_{eff}	Effective refractive index
θ	Angle between optic axis and direction of propagation
E_x	Amplitude of incident beam along X-axis
E_y	Amplitude of incident beam along Y-axis
E'_x	Amplitude of transmitted beam along X-axis
E'_y	Amplitude of transmitted beam along Y-axis
$J(A)$	Jones vector for analyzer

$J(NLC)$	Jones vector for NLC medium
$J(P)$	Jones vector for polarizer
ψ	Angle between optic axis and X-axis
$Q_{\alpha\beta}$	Tensor order parameter
σ	Total stress
g	Intrinsic director body force
N	Angular velocity of the director
A	Rate of the deformation tensor
p	Pressure
I	Unit tensor
γ_1	Rotational viscosity
γ_2	Irrotational torque coefficient
β	Lagrange multiplier vector
$\alpha_1, \alpha_2 \dots \alpha_6$	Six Leslie viscosity coefficient
r	Position vector
v	Velocity

ρ	Density
f	Applied body force
F_d	Free energy density
F_{ext}	External force
ρ_I	Moment of inertia per unit volume
G	Intrinsic director body force
ξ	Director stress tensor
W	Rate of spin tensor
θ_L	Leslie angle
u_x	Flow velocity
$\dot{\gamma}$	Shear rate
Q	Flow rate
d	Channel depth
w	Channel width
K_{11}	Splay elastic constant
K_{22}	Twist elastic constant

K_{33}	Bend elastic constant
I_t	Transmitted light intensity
e	Transition layer of thickness
E	Amplitude
χ	Angle between the polarization directions of the polarizer and analyzer
ϕ	Angle between D' and polarizer
I_0	Input intensity
k	Wave vector
k_1	Wave number
e	Extraordinary wave
o	Ordinary wave
ω	Angular frequency
δ_y	Phase difference along Y-axis
δ_x	Phase difference along X-axis
$f_{electric}$	Applied force due to electric field
V	Applied voltage

V_{th}

Threshold voltage

L_1 and L_2

Lenses

R

Retardation

List of Abbreviations

Abbreviation	Description
CaCl ₂	Calcium chloride
5CB	4-pentyl-4'-cyanobiphenyl
CCD	Charge-coupled device
CMC	Critical micellar concentration
CMOS	Complementary-metal–oxide semiconductor
CPC	Cetylpyridinium chloride
CTAB	Cetyltrimethyl ammonium bromide
DMOAP	Dimethyloctadecyl [3-(trimethoxysilyl)propyl]ammonium chloride
HC-PCF	Hollow core photonic crystal fiber
KI	Potassium iodide
LC	Liquid crystal
LC-PCF	Liquid crystal infiltrated photonic crystal fiber
LCTF	Liquid crystal tunable filters
LOC	Lab-on-a chip

LPG	Long period grating
LPS	Lipopolysaccharide
MEMS	Microelectromechanical system
NaBr	Sodium bromide
NaCl	Sodium chloride
NLC	Nematic liquid crystal
OTMS	Octadecyltrimethoxysilane
OTS	Octadecyltrichlorosilane
PBG	Photonic bandgap guidance
PCF	Photonic crystal fiber
PDMS	Polydimethylsiloxane
POE	Polarization optical element
SAM	Self assembled monolayer
SC-PCF	Solid core photonic crystal fiber
SOP	State of polarization
SPR	Surface plasmon resonance
TIR	Total internal reflection

m-TIR

Modified total internal reflection

XRD

X-ray diffraction

Chapter 1

Introduction

This chapter presents a general introduction to the thesis. The chapter begins with a background of the research problem undertaken. A brief history of optofluidics and how it evolved over the years (after 90's) is given. The requirement for novel optofluidic devices and how a different tuning technique can be achieved by including liquid crystalline properties are discussed. The motivation, objectives and scope of the thesis are discussed in the subsequent sections. Based on the current literature, the proposed and identified research problems are illustrated and detailed with a research road map.

1.1. Background

The term *Optofluidics*, coined in 2003, refers to a class of optical devices fabricated by integrating optics and microfluidics. The fundamental aim is the manipulation of fluid-light interactions and thus developing highly versatile micro systems.

The first developed microfluidic device was the gas chromatography system of Terry *et al.* in 1970's [1]. Inkjet printing is another commercially successful technology that relies on the principles of microfluidics. During the early 90's, researchers across the globe started developing microfluidic devices with properties required in applications such as molecular biology, clinical diagnostics, and cell sorting [2]. The main benefit of scaling down the dimension is that the liquid flow is more controllable in the micron-scale as compared to the macro-scale. The fluidic system has numerous other advantages such as smooth fluid interface, diffusion, excellent transport medium and higher response speed due to small volume.

The incorporation of optics into microfluidics brings additional advantages to optofluidic devices over photonic or microfluidic devices individually. Many of the light-matter interactions such as fluorescence, phosphorescence, scattering, refraction, stimulated emission and polarization can be used for sensing applications. Fast response speed, high sensitivity and localization of light to a scale of few hundreds of nanometers makes these mechanisms ideal for chemical and biological sensing. Thus optics and microfluidics can be combined to either tune the optical properties or to

analyze the fluid characteristics. The main merits of optofluidics are reconfigurability and tunability by the manipulation of fluids. Other advantages are easy fabrication processes (compared to solid-state counterpart/conventional bulk optical systems), low cost and adaptability. Devices, developed successfully using this technology include waveguides, lenses and sensors [3-5]. With advanced fluid handling techniques, it is possible to incorporate all functions in a single chip leading to the successful invention of *lab-on-a-chip* technology. The major requirements of these devices are compactness, high degree of integration and improved functionality. An optofluidic system, incorporating multiple functions in a single chip is shown in Fig 1.1 [6].

Figure 1.1. Illustration of an optofluidics chip [6].

1.2. Motivation

An optofluidic optical component can be defined as a micro fluidic system, operating by controlling its geometry, refractive index and fluid-light interactions. Recently, there is a growing interest in the design and fabrication of tunable optical devices due to their potential applications in biotechnology, sensors and medicine [6, 7]. It is possible to obtain a tunability of $\Delta n \approx 0.5 - 1$ using optofluidics, while electro-optic and thermo-optic tuning methods can achieve, $\Delta n \approx 10^{-3}$ and $\Delta n \approx 10^{-2}$ respectively [8, 9]. The existing methods for refractive index modulation do not provide adequate tuning required at the micrometer scale. *Therefore, novel methods of refractive index tuning need to be developed.*

A major challenge in developing optofluidic elements is the correct choice of fluid. A variety of fluids are used depending upon the applications. However, possibilities of employing an anisotropic fluid in optofluidics are not well explored. Replacing the commonly used fluids with an anisotropic medium provides an additional degree of freedom in these devices.

Liquid crystals (LC), an anisotropic fluid have been studied for more than a century due to its fascinating properties [10]. Therefore a plethora of liquid crystal devices have been developed including displays, polarization optical elements, resonator *etc.* As the name implies, liquid crystals possess crystalline nature even in the fluid state and it is possible to change the material properties by applying an external stimulus.

In majority of existing liquid crystal based optical components, the external perturbation applied is electric field. Owing to the elastic properties of liquid crystals, application of a mechanical force can also generate a change in the characteristics such as refractive index. This concept, which is yet to be explored thoroughly, can integrate with the existing microfluidic techniques to develop a tunable optofluidic polarizer.

Among numerous fluid-light interactions, optical polarization plays a vital role in various systems such as polariscopes, optical switches, variable optical attenuators, sensors, and microscopic image enhancement (such as in polarimetric, hyper spectral and fluorescence imaging). Therefore, it is of utmost interest to tune the state of polarization (SOP) of the optical beam by various technologies. Using microelectromechanical systems (MEMS) and liquid crystal technology, there are reports on the fabrication of optical components for changing the SOP [11-16]. The main concerns in MEMS-based systems are optical losses and complex fabrication processes. In electric field driven liquid crystal devices, the above-mentioned drawbacks of MEMS technology can be subdued. There are still many disadvantages such as low response speed and temperature sensitivity in LC-based systems.

Based on the above considerations, the thesis focuses on investigations of liquid crystal based tunable polarization optofluidic devices with the targeted objectives listed below. The most important aspects are to look into the potential of liquid

crystals and exploring novel refractive index tuning methods for the fabrication of optofluidic devices.

1.3. Objectives

From the previous sections, it can be inferred that the realization of novel optofluidic polarization devices is a research thrust area. The thesis aims to explore a new methodology of achieving tunability by incorporating liquid crystals into the optofluidics system.

In this context, the major objectives of the research thesis can be summarized as follows:

- Fundamental research into liquid crystal-light interactions and their applications as optofluidic tunable polarizer and attenuator.
- Investigations into the effect and dynamic behaviour of thermotropic and lyotropic liquid crystalline materials in an optofluidic platform.

The following section outlines the scope of the research work carried out to meet these desired objectives.

1.4. Scope

Liquid crystals, owing to their elastic properties, can generate a flow induced molecular realignment, upon the application of an external shear force [17-19]. Thus, it can alter the propagation properties of light beam guided through the medium.

Hence there is no need of electric or magnetic field to tune the liquid crystal properties as in the case of conventional approaches, reported in literature [13, 15, 16, 20, 21]. Further, liquid crystal molecular reorientation can change the optical polarization of the light beam traversed through it. Thus, the combination of liquid crystal properties and microfluidic techniques can offer a powerful approach for polarization tuning at the micron level. In this perspective, in order to achieve the major objectives outlined in the earlier section, scope of the thesis can be described as follows.

- Investigations into the fluid-light interactions based on the absorption, scattering and polarization in non-aligned liquid crystalline materials such as lyotropic and thermotropic liquid crystals.
- Design and development of a liquid crystal tunable polarizer and attenuator with flow induced phase shift.
- Fundamental analysis on the molecular dynamics of aligned thermotropic liquid crystals using conoscopy.
- Demonstration of optical polarization tuning due to the liquid crystal molecular reconstruction.
- Experimental validation and respective analysis of the theoretical framework.

A detailed research road map outlining the research methodology executed for this thesis is given in the flow chart [Fig 1.2].

Figure 1.2. Research road map.

1.5. Organization of the Thesis

The thesis is divided into 5 chapters following the introductory chapter. Each chapter begins with a short abstract, which reflects the main contents of that chapter.

Chapter 2 reviews, work done in the related areas of optofluidic devices with a detailed analysis of various tuning methodologies that are reported in the literature.

For using liquid crystal for the fabrication of polarization optical elements, the existing techniques employed in existing devices are also discussed. A general review of various technologies used for the design and fabrication of tunable polarizer and a comparison of different techniques are also given in this chapter. The outcome of literature review is discussed briefly towards the end of the chapter.

In *Chapter 3*, fluid-light interactions in a non-aligned lyotropic liquid crystal under flow are analysed. The effect of a lyotropic liquid crystalline material, cetyltrimethylammonium bromide (CTAB), a cationic surfactant in changing the polarization axis of the input light is analysed in detail. Here the liquid crystalline phase formation in CTAB below Krafft temperature is analysed. The effect of Poiseuille flow in varying the SOP is investigated followed by the contribution of absorption and scattering.

Chapter 4 begins with a brief introduction to thermotropic liquid crystalline materials and mainly focuses on the flow induced variations of the same. The effect of flow in changing the polarization is analysed. Beam fanning, a scattering effect occurring due to the refractive index modulation is monitored and analysed in detail. Further, the contribution of absorption in the transmitted intensity is explored as well. Thus chapter 3 and 4 discusses the possible optical interactions with different kinds of non-aligned liquid crystals under flow.

Chapter 5 illustrates the dynamics of aligned thermotropic liquid crystal. Different alignment methods used in the fabrication of liquid crystal cells are

discussed briefly. The molecular reconstruction occurring inside the glass microchannel with respect to flow is studied using conoscopy, an interferometric method. The refractive index variation due to flow in the NLC medium is analysed through optical transmittance studies. Thus chapter 5 demonstrates the possibilities of using the flow driven nematic liquid crystal medium as a tunable polarizer.

Chapter 6, the last chapter, gives the conclusion for the thesis along with the guidelines for future work. It explains the important research contributions made in this thesis. All the theoretical and experimental works associated with the proposed research for fulfilling the required objectives are concluded here. Also, certain suggestions are provided for extending the application areas of the proposed system by incorporating additional elements.

Chapter 2

Literature Review

The chapter reviews the earlier work done in areas that are relevant to the thesis. The chapter begins with the descriptions on various tuning techniques adapted for optofluidic devices. Subsequently, information about the integration of multiple functions in a single chip exploring the potentials of optofluidics is also presented. With a view to considering liquid crystal for the fabrication of polarization optical elements, the existing techniques employed for the same are also discussed. Also, it provides a general idea on different types of liquid crystals. The chapter is concluded by stating the outcome of the literature review.

2.1. Introduction

Majority of the optical components are fabricated using glasses, metals and semiconductors. However, there were certain systems made with the inclusion of fluids such as dye lasers, oil immersion microscope and liquid mirror telescopes long time ago [22-24]. But these technologies have not been much explored in the micro-scale. With the development of microfluidics during early 90's, researchers started to work on fluidics-based optical devices in the following years. Successful invention of such devices resulted in the development of a new research field known as *Optofluidics*. Generally, optofluidic devices are classified based on their application or mode of operation which are mainly two fold. Optical properties can be used to control fluid motion and also for its characterization, with main applications in the field of bio sensing. Conversely, fluids can be exploited for the fabrication of various tunable and reconfigurable photonic devices as illustrated in Fig 2.1.

Figure 2.1. Classification of optofluidics devices based on the operation.

A brief description on various techniques and different optofluidic devices is illustrated in the following sections.

2.2. Optofluidics with Light Manipulating Fluid

Light can trigger the motion of fluid or can control small particles immersed in the fluid by different methods. This can be achieved by heating, applying pressure and also using a photo responsive surface. Motion control using heating is achieved by thermo capillary phenomenon known as *Marangoni effect*. Kotz *et al.* demonstrated the movement of aqueous droplets, immersed in decanol using laser heating. The rate of movement of droplets was reported to be 3 mm s^{-1} [25].

Later, Liu *et al.* reported laser controlled microfluidic flow by heating the nanoparticles immersed in the fluid [26]. Heat generated in the photothermal particles causes the fluid to evaporate and condenses into small droplets making the surface wet. This method can achieve a flow rate of $500 \text{ } \mu\text{m s}^{-1}$. The movement of fluid molecules using pressure was achieved by exposing the surface to electromagnetic radiation. There have been reports based on the movement of desired biomolecules by light scattering forces, a suitable method for cell sorting applications [27]. The movement of liquid droplet using a photoresponsive surface was also demonstrated [28]. An isomerizable compound isobenzene was coated on the surface and the irradiation of the surface triggers the motion of droplets due to the phenomenon known as photoisomerization. The direction and speed of the movement

was controlled by the light intensity gradient. Photographs and schematic of the movement of olive oil droplet is shown in Fig 2.2.

Figure 2.2. An illustration of surface driven motion of droplet using the process of isomerization [21].

Fluidic analysis using integrated photonic structure consisting of nano arrays was reported by Leebeeck *et al.* in 2007 [29]. Chemical sensing was achieved by monitoring the transmission spectra from the nanohole array. Another group of researchers reported a similar optofluidics sensor. The sensing method was based on surface plasmon resonance (SPR) from nano hole arrays in the metal [30]. Surface enhanced Raman scattering phenomenon was also used for the same [31].

2.3. Optofluidics with Fluid Manipulates Light

Adapting different properties of fluids, it is possible to fabricate different optofluidic devices such as lasers, lens, mirrors, attenuator and optical tweezers. Based on the method to achieve required tunability, optofluidic devices can again be sub-divided into two groups as shown in Fig 2.3. Tunability in optofluidic devices can be achieved either by geometric variation or refractive index modulation.

Figure 2.3. Different techniques used in optofluidics to achieve tunability.

2.3.1. Optofluidic Devices with Geometric Tuning

Optofluidic device performance often depends on the geometry of the boundary of the device such as liquid-liquid, solid-liquid, liquid-gas and solid-gas [32]. The shape of the optical interface can be changed by the application of pressure or external forces, electric field (electro wetting), and optical trapping

(optical tweezers). A brief description on optofluidic components with tunability achieved geometrically is given in the following section.

In the fabrication of most of the optofluidic devices, soft elastomer polydimethylsiloxane (PDMS) is used. The elasticity of this material was utilized for tuning the optical properties by applying pressure. Li *et al.* demonstrated an optofluidic distributed feedback dye laser in which one of the methods of tunability was by stretching or compressing the PDMS chip [33]. Thereby, the periodicity of the distributed feedback structure was changed. The schematic diagram of the tunable laser is shown in Fig 2.4.

Figure 2.4. Schematic diagram of mechanically tunable distributed feedback dye laser. Upper inset shows the PDMS laser chip and lower inset is the optical micrograph image of phase shifted region in the laser cavity [33].

A tunable lens was reported by deforming a membrane made up of PDMS. The membrane was filled with a fluid and the shape of the membrane was varied by the application of pressure [34, 35]. Silicone tubes were connected to the fluid reservoir using glue and suitable pressure was applied on the fluid by means of a syringe pump. Depending upon the pressure, shape of the PDMS membrane changes and thereby varying the radius of curvature [35]. An illustration of the tunable lens is shown in Fig 2.5.

Figure 2.5. Tunable microfluidic lens reported by Werber *et al.* [35].

A variable optical attenuator (VOA) was reported, in which the attenuation was controlled by changing the optical transmission path of the input beam [36]. Controlling the amount of light absorbing liquid in the optical path, the desired

attenuation was obtained. This was achieved by the deformation of a membrane made up of PDMS and obtained an attenuation of 38 dB [Fig 2.6].

Figure 2.6. Variable optical attenuator reported by Hongbin, *et al.* [36].

Varying the wetting angle by the application of a electric field (electro wetting) was another method for achieving tunability geometrically [37]. The schematic of the tunable lens is shown in Fig 2.7.

Figure 2.7. Schematic of electric field driven tunable mirror [37].

The system consists of a droplet of conducting fluid, in contact with an ITO electrode. In the presence of voltage, the surface of the droplet was deformed and became a highly reflective flat surface to function as a mirror. Due to the spherical shape of the droplet, the device can also be used as a negative lens. A tunable micro mirror was also reported based on electro wetting [38]. This was done by changing the curvature of the interface between the immiscible fluids. The shape of the spherical droplet of the conducting fluid was changed by applying voltage. Thus the focal length of the mirror was tuned as depicted in Fig 2.8.

Figure 2.8. Change in the curvature of two immiscible liquids by the application of voltage (a) with no voltage applied, the curvature of the droplet is spherical (b) With voltage applied, curvature is changed with positive focal length [38].

A different approach other than electro wetting for achieving tunability is using optically trapped particles. Optical trapping or optical tweezing is a non-invasive technique to move micro objects in liquid or gas medium. This has been widely used in handling biological cells. This method of manipulating dielectric

particles using strong electromagnetic energy was first demonstrated in 1980's [39]. It is possible to trap particles of size up to 20 microns using high power laser beam and can be manipulated by changing the focus of the trapping beam [40]. The same method can be used in optofluidics to change the transmission properties. In a report by Domachuk *et al.*, the intensity of the input beam was controlled by changing the position of a trapped micro sphere dispersed in water in a microchannel [41]. This configuration can function as a beam steerer and attenuator. Also the above system can be used for fluid manipulation and detection.

Figure 2.9. Diagram representing the tunability achieved in light transmission by optical trapping method [41].

2.3.2. Optofluidic Devices with Refractive Index Tuning

Tunable optofluidic devices are also fabricated by varying the refractive index of the medium. This includes replacing the fluids or mixing of fluids, changing the fibre properties, doping with foreign particles, applying external perturbations

(electric field, temperature and mechanical force) etc. Commonly used fluids in optofluidics technology are distilled water, solutions of calcium chloride (CaCl_2), sodium bromide (NaBr), potassium iodide (KI), glycerol, sodium chloride (NaCl), ethanol, rhodamine 6G and methylene iodide.

One of the simplest methods for refractive index modulation is by the exchange or mixing of fluids of different refractive indices. A variable optical attenuator was reported by Zhu *et al.*, consists of a waveguide integrated with a fluidic channel [42]. The refractive index of the cladding layer was varied from 1.557 to 1.584 by switching between two solutions and an attenuation of 28 dB was achieved. After switching from one solution to another, there can be traces of previously used solution and this can affect the performance of the device. In addition to the exchange of fluids, refractive index modulation can also be achieved by mixing of different fluids. A tunable optical filter consisting of a micro ring waveguide resonator inside a microchannel was reported, utilizing the diffusion property of fluids as shown in Fig 2.10 [43]. Fluids introduced into the channel, acted as the upper cladding layer of the waveguide. Transmission through the waveguide was controlled by the refractive index variation of the cladding layer by mixing different solutions.

Figure 2.10. Schematic of an optofluidic tunable filter [43].

Galas *et al.* reported another optofluidic device using refractive index modulation, a tunable laser [44]. Tunability in the emission wavelength was obtained by changing the concentration of gain medium by mixing with ethanol. Using the same approach of mixing of solutions, a variable optical attenuator was also reported [45]. Here, the incident light was focused into the microchannel, filled with a fluid. The light reflected from the interface between the fluid and the channel, was collected by an optical fiber. The desired attenuation was achieved by varying the refractive index of the fluid flowing through the channel by adding other suitable fluids into it. Thus optical reflectivity was controlled. In this case, the refractive index was varied from 1.33 to 1.414 and thus attained an attenuation of 35 dB. The major drawback with this type of refractive index modulation is the complex mixing networks. Schematic of the variable optical attenuator is shown in Fig 2.11.

Figure 2.11. Variable optical attenuator using the principle of Snell's law of refraction [45].

Recently optofluidic components were fabricated by manipulating the properties of microstructured optical fibers by introducing fluids into it. In the optical filter reported by Kerbage *et al.*, a long period grating (LPG) was engraved in the fiber core. Low index and high index fluids were injected into the cladding layers adjacently, to act as a waveguide [46].

Figure 2.12. Tapered microstructured optic fiber with fluid plug infused into one of the cladding layer [46].

The propagation characteristics of the fiber can be tuned by changing the position of the microfluidic plugs in the cladding layer. An attenuation of 12-15 dB was obtained using this method.

Polarization tuning based on optofluidics was also reported by the same group [46]. Here the medium was a tapered microstructured optical fiber. In this case, birefringence was created in the fiber by selectively filling one of the cladding layers. This breaks the symmetry of the microstructured fiber. Tunability, in the state of polarization was obtained by varying the length of the fluid interacting with the mode field. A capillary heater was used to move the fluid plug by inducing pressure. Using the above configuration polarization axis can be rotated by 2π , moving the fluid plug along 4 cm of the fiber.

Figure 2.13. (a) Polarization phase change with respect to the temperature, (b) Relation between the length along which the fluid plug is moved through the fiber and temperature of the capillary heater [46].

Using tapered microstructured fibers, variable optical attenuators was also reported [46]. Here the output light intensity was controlled by introducing a high index fluid (methylene iodide) into the cladding layer. The fluid movement was controlled using a heater. Light gets refracted through the high index fluid. The switching speed was about 0.5 sec and is not sufficient for communication purposes. Since optical fibers are sensitive to temperature, the thermal heater used for moving the fluid plug can change the property of the fiber. Thus the successful operation of the attenuator can be affected. Also, when the fluid was moved from waist of the fiber for the maximum transmission of light, traces of the fluid can affect the operation.

Another method of achieving the refractive index modulation is thermo-optic effect. In 2006, Tang *et al.* reported an optical waveguide with transmission controlled by thermal effect [47]. Fluids of different refractive indices act as core and cladding. The refractive indices of core-cladding layers were maintained by applying different temperature. Refractive index of most of the liquids decreases as temperature increases. Therefore, in this configuration, fluid at higher and lower temperature acts as the cladding and core, respectively [Fig 2.14]. Refractive index modulation using thermal effect was also mentioned by doping the fluid with suitable nano particles. Optical and thermal effects occurring in a colloidal solution of silver nano particles was reported by J. R. Adleman *et al.*, using holographic method [48]. The thermal energy absorbed by the nanoparticles, heat the surrounding solvent and thus the refractive index was changed. This phenomenon is known as Soret effect

(thermophoresis). Silver nanospheres of average diameter 3.8 nm, dispersed in toluene were used.

Figure 2.14. (a) Schematic diagram of thermally controlled waveguide (b) Variation of refractive index with respect to temperature [47].

Electro-optic method for obtaining the variation in refractive index can be done by utilizing the properties of liquid crystalline (LC) materials which is not well explored in the field of optofluidics. A liquid crystal based beam deflector was reported by Wang *et al.* [49]. The device operates by modifying the refractive index of the liquid by the application of voltage. The change in the refractive index of the material by applying electric field is due to the birefringence property of liquid crystals [Fig 2.15].

After the successful development of optofluidics, researchers found the potential of incorporating all the functions into a single chip creating lab-on-a-chip devices [50]. All the above mentioned optofluidics devices can be integrated to form

such devices. A brief description on the fabrication of on-chip devices are given in the next section.

Figure 2.15. Schematic representation of LC based beam deflector [49].

2.4. Lab-on-a-Chip Devices

Lab-on-a-chip (LOC), is a miniature form of a lab for microanalysis in a single substrate [51]. A variety of optofluidics devices and tuning methods are beneficial for LOC devices. Microfluidics is the basic toolkit for the LOC device fabrication and the invention of optofluidics can be envisioned to bring large impact on this field. Optofluidics, opens up new possibilities for control of fluid flow in microchannel, optical trapping and biosensing [26, 41, 52, 53]. Recently, Balsev *et al.* reported a lab-on-a-chip system integrating optics, fluidics and electronics [54]. The device includes a dye pumped laser, waveguides, diffusive mixers and photodiodes. The light emitted by the laser can be transmitted into five different locations through the

waveguides along a channel and the transmitted light was detected using the photodiodes. Thus the LOC device acts as a biosensor.

Figure 2.16. An optofluidic lab-on-a-chip device [54].

In the field of sensing, the main advantages of optofluidic LOC devices are,

1. Fluid is an excellent medium for biological samples and control of the flow provides an easy method of transport and delivery of samples.
2. Optical sensing method is fast compared to fluidics techniques and variety of optical sensing methods are available [55, 56].
3. Micro and nanochannels require only small sample volume.

Recognizing the advantages offered by optofluidics, many of the optofluidic devices are already miniaturized as a chip. One of the important applications of optofluidics is to control the movement of micron-scale objects and thus acting as optical tweezers. Cran-McGreehin *et al.* demonstrated optical tweezers with on-chip

optical path and is a very interesting LOC device [57]. Another important LOC device, is an optofluidic microscope which provides on-chip imaging as shown in Fig 2.17 [58].

Figure 2.17. Schematic representation of an optofluidics microscope [58].

The sample was allowed to flow through a microchannel with a series of apertures over a CMOS layers so that it can be imaged. The image resolution was controlled using the aperture size. In future, this invention can be very successful in miniaturizing bulk optical systems into compatible ones. LOC devices mainly focus on biomedical spectroscopy, imaging and sensing. Voltage driven liquid crystal devices such as tunable filters and waveplates have been included in many of the spectroscopic and imaging techniques. The main potential of integrating liquid crystals into these systems is to improve the accuracy and increase the system functionality. The high sensitivity of liquid crystals to many of the biological samples

makes it beneficial in the field of sensing. Therefore, the subsequent section illustrates how the liquid crystal can be used for the same.

2.4.1. Liquid Crystal for Spectroscopy, Imaging and Biosensing Applications

Spectroscopy and various types of imaging are widely used tools in biomedical applications such as medical diagnosis and research. Liquid crystal based tunable filters (LCTF) can be used in spectrometers as an alternative to dispersive grating based techniques. The working of LCTF tunable filter is based on changing the optical retardance of the medium with respect to an externally applied stimulus. One of the limitations of LCTF based spectroscopy system is that multiple stacks of liquid crystal cells are needed to obtain the desired retardation. The design of new non-dispersive liquid crystal based filters which is cost effective and compact can result in new possibilities, which earlier been prohibitive due to cost and size. Other than spectroscopy, liquid crystal based devices can be used in hyperspectral and polarimetric imaging. Hyperspectral imaging with LC-based components, ranging from molecular to cellular have been used for mapping of oxygen saturation, assessment of cardiac oxygenation and endoscopic diagnostics [59-61]. These applications have just started to flourish. As discussed earlier, the improvement in the current field can push the applications of hyperspectral imaging to various biomedical areas which are not yet explored.

Polarimetry and polarization microscopy is yet another field used for the analysis of the materials of interest based on the polarization state of light. For

characterizing liquid crystal materials often polarimetry is used, but it is also possible to integrate liquid crystal based components for the device operation. The birefringence nature of many of the biological specimens makes polarimetry an effective method in bio-applications. Liquid crystal has been incorporated into polarization microscopes by many researchers as polarization state generator (capable of making different polarization states) and polarization state analyzer (used to analyze the output beam polarization state) [62, 63]. There has been an effort to miniaturize the same which includes microcell polarimeter, integrating liquid crystal in an optic fiber [64]. An in-line optic fiber polarimeter can be used either to analyze the polarization properties of the fiber or to collect the specimen interacted light. Liquid crystal based polarization modulators and phase shifters can improve the performance and image collection speed in polarization microscopy. Fast response speed of LC-based components also contributes in birefringent imaging microscope which gives more structural details [65, 66]. Here a stack of liquid crystal variable retarders, driven by an external voltage was used to act as the optical compensator. One of the main advantages of polarimetry and polarization microscopy is that there is no need of dyes, fluorescent labels etc and is less destructive when compared to other techniques such as electron microscopy. The applications of liquid crystal in spectroscopy and microscopy are quite wide, still there are many more to explore. The current applications of liquid crystal tunable filters in spectroscopy and other imaging techniques show that, biomedicine has benefitted by this technology.

Liquid crystal also provides a means to characterize the biological samples and is an excellent medium for biosensing applications. Generally many of the biological samples resemble the characteristics of lyotropic liquid crystalline materials. Thermotropic liquid crystals are used as the substrate for the lyotropic biological samples. When the biological samples are made in contact with a thermotropic liquid crystal interface, the orientation prevailed in the bulk liquid crystal medium changes. This provides the basis of liquid crystal biosensors. Therefore, the major objective in this direction is to identify the various ways in which biomolecules interact with the liquid crystals.

Brake and colleagues has studied the effects of phospholipids at the interface of aqueous liquid crystal, in changing the orientation [67]. The interface of liquid crystal with an aqueous medium induces a planar alignment and the addition of lipids induces a change in this orientation. Fig 2.18 demonstrates a schematic of the alignment changes with respect to time. The effect of an enzyme, which converts lipids into fatty acids in changing the orientation, was also demonstrated. This provides a method to monitor the enzymatic action using a phospholipid decorated liquid crystal surface. Liquid crystal biosensors can also be used to detect the protein binding events. The main aspect of this application is the disruption of liquid crystal molecular alignment due to the immobilization of proteins on the surface [68].

Figure 2.18. Polarizing microscope images and a schematic representation of changes in the alignment of liquid crystal molecules in the presence of phospholipids (a) immediately (b) after 20 minutes (c) after 2 hours [67].

Espinoza *et al.* demonstrated the orientation of nematic liquid crystal in contact with a virus [69]. It was shown that with time, the molecular alignment changes from planar to homeotropic on the virus bound surface. Also it is possible to identify the external structure of virus based on the induced alignment changes of nematic liquid crystal molecules [70]. Here a cell was fabricated with virus contacted and octadecyltrichlorosilane (OTS) treated surfaces as the bottom and top substrates, respectively. Later a nematic liquid crystal was drawn into the cell using capillary action and optical images were taken using polarization microscopy. The experimental results were also confirmed using the method of conoscopy. Another

application of liquid crystal in medical diagnosis is the detection of bacteria in bio-specimens. McCamley *et al.* demonstrated that, Gram negative bacteria induces an orientational change in a liquid crystal medium [71]. The long chained molecule (lipopolysaccharide, LPS) present in the bacteria is the reason for this change. This was validated by conducting experiments with different concentration of LPS in contact with liquid crystal medium. Even though recently there are reports on liquid crystal based biosensors, still the current field is in its infancy. One of the challenges in this field is the liquid crystal biocompatibility, which has to be ensured for the successful operation of a sensor. To analyze the main mechanism of alignment changes occurring in liquid crystal molecules in the presence of biomolecules, it is of utmost importance to understand the effects of liquid crystal on biospecimens. Other key factor is the proper design of a cell to carry the biomaterials and liquid crystal.

As indicated in chapter 1, employing anisotropic fluids in optofluidic devices has not received much attention. In the subsequent sections, the various possibilities of a tunable optofluidic polarizer based on the laminar flow of liquid crystals are discussed. By controlling the pressure driven flow, liquid crystal properties can be manipulated. Details of the methodology adapted in the thesis and a brief description on different types of liquid crystals are given in the following section.

2.5. Liquid Crystal Based Optofluidics

Generally there are two types of liquid crystalline materials, namely lyotropic and thermotropic. Lyotropic compounds consist of amphiphilic molecules and various

liquid crystalline phases are observed depending upon parameters such as concentration, temperature and addition of salts. In thermotropic LC, the liquid crystalline phase transition takes place as a function of temperature [72]. More details regarding lyotropic and thermotropic LC are given in chapter 3 and chapter 4, respectively. In LC materials, molecules vary spatially or get deformed upon the application of an external perturbation. Due to the large birefringence, as the LC molecules reorient, their optical properties are also changed and thus can be used for varying the state of polarization.

In earlier section of this chapter, a brief discussion on different optofluidic devices is given. Among them, a polarization optical element based on an optofluidic platform has not been much explored. A polarization optical element (POE) changes the state of polarization of the incident light beam such as polarization rotator, variable phase modulator, wave plates and polarization controller. Polarization rotators are used to rotate the orientation of an incident-linearly-polarized light still maintaining the linear polarization at the output. A phase modulator changes the phase of the transmitted optical beam, thereby changing the polarization. Wave plates can be utilized in rotating the plane of polarization and also induce a phase shift (retardance). Polarization controller converts an arbitrary polarization into a known polarization. Therefore, it can be seen that the main application of all the above mentioned polarization optical elements is the tuning of optical polarization in different ways. Nowadays these optical elements gain much importance because of

its potential applications in spectroscopy, sensing, optical switches, polarimetry and variable photonic delay line modules. A brief description of different methodologies adopted for polarization tuning, microelectromechanical systems (MEMS) and liquid crystal based technologies are illustrated in the following section.

2.5.1. MEMS Based Tunable Polarization Optical Elements

Pu *et al.* reported a MEMS based polarization rotator, in which the required phase shift was obtained by varying the optical path difference [11]. The system consists of beam splitters and micro mirrors on a single silicon chip. One of the micro mirrors was attached to an electrode plate and the optical path difference was changed by varying the position of the micro mirror applying a bias voltage to the electrode. Here a phase shift of 2π was achieved at a bias voltage of 45 V.

Figure 2.19. Schematic representation of MEMS based polarization rotator [11].

Another mirror based polarization rotator, replacing the need of polarization beam splitter (PBS) configuration was also reported [12]. The configuration consists of four

mirrors, of which two mirrors were fixed (M2 and M3). Depending upon the position of the active mirror M1 with respect to the incident light beam, the optical path can be varied as depicted in the Fig 2.20. The main concerns with MEMS based system are complex configuration and optical loss.

Figure 2.20. Schematic representation of polarization rotator with optical path directed along (a) Y-axis (b) X-axis depending upon the position of M1 [12].

2.5.2. Polarization Tuning Using Birefringent Materials

A further approach of changing the state of polarization was using the birefringent properties of anisotropic materials such as lithium niobate, barium titanate and liquid crystals. The basic principle of polarization rotation is the phase modulation achieved through externally-controlled birefringence. Among POE using different anisotropic materials, the LC based devices are the most mature and commercially successful technology.

LC based POE usually consists of several wave plates and LC cells, stacked together with the voltage applied to each of the cell. Polarization rotator, consists of a linear polarizer and two LC phase retarders kept at 45° with respect to the other was reported [13]. Here the optic axis was rotated from 0° to 90° by applying voltage to the LC cell.

Figure 2.21. LC polarization rotator consists of one polarizer and two phase retarders [13].

In another work, a twisted nematic LC was kept between two quarter wave plates and the polarization axis of the incident light was changed by varying the phase, as a function of applied voltage [14]. LC based linear polarization rotator was also reported by Safrani *et al.* in which two LC retarders were placed between the quarter wave plates. Here, the plane of polarization was rotated by 90 degree with an increment of 5 [15].

Figure 2.22. Schematic of LC based tunable polarizer [15].

The polarization axis rotated from 0° to 90° by rubbing method is another approach [16]. The cell filled with LC material was rubbed in such a way that the molecular orientation follows a twist deformation along the cell. The LC molecular deformation with respect to the rubbing direction is shown in Fig 2.23. In this way, polarization axis was continuously rotated by changing the position of the LC cell

with respect to incoming light. Here any misalignment can cause serious errors in the expected result.

The polarization devices based on liquid crystal materials has many advantages such as low optical loss, no moving parts and compact size when compared to MEMS technology. Even though there are merits, some of the drawbacks of LC based systems are low response time and fluctuations due to temperature dependence. Therefore, there is a need to develop novel methods for tuning the state of polarization.

Figure 2.23. Schematic representation of LC based polarization rotator based on the rubbing method [16].

2.6. Outcome of Literature Review

The research and development of optofluidics, a multidisciplinary field, has gained significant progress during the recent years. Optofluidics has enabled the integration of multiple fluid tasks and various optical processes in a single chip. Compared to electronic methods, optical detections systems are more preferred due to

high sensitivity, less chance of electromagnetic interference, and non-contact detection. Therefore, optofluidics have gained considerable attraction for lab-on-a-chip applications and it automatically encourages researchers to explore novel methods for the realization of the same [54].

Apart from the possibilities in the field of sensing, optofluidics also provides a way to tune microphotonic devices [6]. In addition to the optical tunability achieved using the elastic property of the constructing material, large group of optofluidics devices rely on tunability enabled by fluid properties. Thus, in optofluidic devices the tuning methods can be divided into two groups: refractive index tuning and geometric tuning. Since most of the optofluidic devices are fabricated using a soft elastomer, by applying an external force, the shape of the device can be changed [33, 35]. Thus the required tunability can be attained geometrically. The usual methods adopted to achieve refractive index tunability are by the exchange and /or mixing of fluids of different refractive indices and by doping with suitable particles [44]. Based on the literature survey, it is also inferred that the polarization tuning in an optofluidic platform has not been studied thoroughly yet. The incorporation of liquid crystal can provide a better way to design a tunable optofluidic polarizer. MEMS and liquid crystal based systems are the two techniques used for tuning the state of polarization. Even though LC based devices have many advantages compared to other methods, still it is difficult to incorporate these systems into LOC devices.

A variety of fluids are used in optofluidic devices depending upon the applications. The inclusion of anisotropic fluids in optofluidic devices has not received much attention. However, replacing commonly used fluids with liquid crystals may offer novel methods of refractive index tuning that can be easily integrated onto a compact chip. Also, optical polarization tuning with respect to liquid crystal flow is currently an understudied area and it is noteworthy in enhancing the photonic device functionality.

In this context, this thesis focuses on these above mentioned aspects to investigate flow induced SOP variation using liquid crystals. It can be inferred that by providing necessary tuning mechanisms, optical properties such as polarization, amplitude and phase can be controlled using liquid crystals. Liquid crystal integration in optofluidics holds much promise for developing polarization based devices and also highly sensitive biosensors. This is expected to have potential applications in the miniaturization of complex laboratory functions.

Chapter 3

Investigations into the Fluid-light Interactions in Lyotropic Liquid Crystal

This chapter describes the effect of a lyotropic liquid crystalline material, cetyltrimethylammonium chloride (CTAB), a cationic surfactant in changing the polarization of the input optical beam. Here, the liquid crystalline phase formation in CTAB below Krafft temperature is analysed. It is found that the lamellar structures formed in the material below Krafft temperature are birefringent and this birefringence can be controlled by flowing it in rectangular channel under a suitable flow rate. The effect of other fluid-light interactions such as absorbance and scattering has also been analysed

3.1. Introduction

Tunable optofluidic components utilizing different fluid properties generated a growing interest in the recent past among the research community. In this research, we used lyotropic liquid crystalline materials. In nature, most commonly found liquid crystals are lyotropic such as cell membranes in the human body. Surfactant is a well-known synthetic lyotropic liquid crystalline material. A surfactant molecule consists of polar hydrophilic group (head part) and non-polar hydrophobic group (tail part). Schematically amphiphilic molecules are usually represented as circles followed by zig-zag lines depicting head and tail part, respectively. The important property of lyotropic liquid crystal is the formation of micelles when it is dissolved in suitable solvents. Micelles are formed when the hydrophobic tail part aggregates away from the solvent by surrounding the hydrophilic head group around themselves.

In aqueous solutions, there is a critical concentration above which the micelles start to form, known as critical micellar concentration (CMC). Above CMC, these molecules will form different types of aggregates such as spherical micelles, disk shaped, wormlike or rod like micelles, and reversed micelles. Reverse micelles are usually formed in water-oil mixtures, where water content is less. Water reside inside the head part rather than outside, as in the case of ordinary micelle. The growth of aggregates into various shapes depends on conditions such as concentration of surfactant, temperature, presence of salts and mixing of surfactants.

As mentioned above, one of the parameters that play a crucial role in the crystalline characteristics of surfactants is temperature. It is noted that the properties of aqueous solutions of surfactants above and below a particular temperature known as *Krafft temperature* is entirely different. Above this temperature, surfactant is highly soluble and the micelles start to form. Various theoretical formulations are established to explain the change in the morphology of aggregates [73]. The properties of aqueous surfactant solution such as rheology and flow birefringence, above the Krafft temperature has been studied extensively [74-84]. Flow birefringence, upon the application of a shear force in a surfactant solution has been studied in detail. These have been demonstrated in various ways such as in simple shear flow, through extensional flows such as cross slot flow cell and in T- shaped channel which is much more complicated [85-91]. However, the properties below Krafft temperature were inadequately investigated [92, 93]. In this context, the birefringence property of a cationic surfactant, cetyltrimethylammonium bromide (CTAB) which is a lyotropic liquid crystalline material below Krafft temperature is studied in detail and discussed in this chapter.

The chemical structure of CTAB is shown in Fig 3.1 [94]. A CTAB molecule consists of a tail (hydrophobic) and head (hydrophilic) part. It is reported that at lower concentrations ($\sim 9 \times 10^{-4}$ to 0.3 mol/l) spherical micelles were formed and in the concentrations range between 0.3 to 0.7 mol/l, rod like particles were observed [95].

Figure 3.1. Chemical structure of CTAB [94].

However, at high concentrations (above 0.7 mol/l), a liquid crystalline phase consisting of long threads was observed [96, 97]. This crystal like behaviour suggests that the micelles have a significant shape asymmetry.

It is reported that when the temperature is below Krafft temperature (25°C for CTAB) the solution exists in a two phase form; hydrated crystals and water [82]. The formations of hydrated crystals are due to the crystallization of alkyl chains (hydrophobic) and the head group (hydrophilic) of the surfactant molecule. The crystallization is favored when the Van der Waals attraction energy exceeds thermal kinetic energy. The formation of hydrated crystals in one of the surfactants, cetylpyridinium chloride (CPC) in aqueous solution was reported [93]. It is described that during the formation of hydrated crystals, a metastable-stable transition takes place. Metastable solids are needle shaped and stable solids are plate shaped crystals. The transition from metastable to stable solids can occur in two ways. The rearrangement of metastable solids into a stable structure by themselves is one of the mechanisms and the second is the solvent assisted process.

3.2. Experiment

3.2.1. Sample Preparation

Cetyltrimethylammonium bromide (CTAB, Aldrich Chem. Co) was used without further purification. Solution was prepared in double distilled water. Initially CTAB solution was kept above Krafft temperature for few hours to get a clear solution. When the surfactant became completely soluble in water, the solution was cooled down to a temperature below 25°C where the hydrated crystals were formed by precipitation. The transparent solution became turbid due to the effect of temperature. This solution was kept for 2 days to attain equilibrium. It was observed that after 2 days, the hydrated crystals got aggregated more and was unable to flow it through the cell. Just before the experiment, the turbid solution was stirred which became glossy after stirring.

3.2.2. Spectroscopic Measurements

In order to characterize the hydrated crystals, absorption spectrum, microscope images and diffraction experiments were carried out. UV-Vis Spectrum was measured using Shimadzu Corp. (UV-1601PC) spectrophotometer and the baseline was corrected with water. Optical microscope images were taken using Carl Zeiss, Inc. (Axiotech) microscope. X-ray diffraction characterization of the sample was done using Philips (PW1830) X-ray diffractometer. Samples were prepared by drop casting of the solution on glass slides.

3.2.3. Fabrication of Flow Channel

A 25 mm long, 2 mm wide and 1 mm deep, rectangular flow channel or flow cell was fabricated using microscopic glass substrate. The schematic of channel fabrication is shown in Fig 3.2 (a) and (b). All sides of the channel were made of glass with a refractive index 1.52. Initially two similar glass slides were mounted parallel on a bottom glass slide. The distance between these two upper glass slides was 2 mm. Then, another glass slide was mounted on the top of these spacers to form the flow channel. The flow cell was fixed by using standard glue (Araldite, an epoxy adhesive). At both ends of the channel reducing connectors were glued to connect to external silicone tubing.

Figure 3.2. Schematic of channel fabrication steps (a) A rectangular channel made using glass slides (b) The channel with inlet and outlet.

The flow cell was connected to a syringe pump using silicone tubing with an outer diameter of 0.6 cm and an inner diameter of 0.3 cm [Fig 3.2(b)]. The channel was cleaned before introducing the surfactant.

3.2.4. Experimental Set Up and Procedure

Schematic diagram of the experimental set up used for the investigation is shown in Fig 3.3. Diode laser with a wavelength of 641 nm was used for the experiment as the source. The output power was monitored using a (Thorlabs, Inc. PM100) power meter which is connected to a computer for data analysis. The flow cell was kept between crossed polarizers to analyze the birefringence of CTAB solution. A linearly polarized laser beam was focused to the flow cell and the transmitted beam is again passed through a second polarizer (analyzer) in which polarization axis is perpendicular to the first one.

Figure 3.3. Schematic of the experimental set up.

CTAB solution was injected into the flow cell using a syringe pump (Cole Parmer, EW-74900-05) with a constant flow rate of 80 ml/hr. The output laser power was monitored for a period of time. Effect of pumping time was also studied. This was done by maintaining a constant rate of flow for different time period (30 sec and 250 sec) and monitoring the variations in the transmitted power.

The transmitted light intensity variation upon changing the CTAB concentration (0.4 M, 0.5 M and 0.6 M) was also monitored. To analyze the birefringence property of hydrated crystals in more detail, experiments were performed with magnetic stirring of the fluid (CTAB solution). A small magnet was placed inside the syringe and actuated by a magnetic stirrer (Favorit-HS0707V2) [Fig 3.4]. The influence of different flow rates on output intensity was also noted.

Figure 3.4. Schematic of the modified experimental setup.

3.3. Results and Discussion

The optical microscopic image of the hydrated crystals formed in CTAB solution is shown in the Fig 3.5. It clearly shows the metastable state (needle shaped) and the stable state (plate like) hydrated crystals. Metastable-stable transition takes place in few hours after the formation of hydrated crystals. Fig 3.6 shows the absorption spectra of CTAB in aqueous solution. The spectrum is characterized with a peak at 216 nm and a broad band between 260 nm to 310 nm. This clearly indicates that there is no absorption in the visible region.

Figure 3.5. Optical microscope image of the hydrated crystals formed in CTAB solution (A) metastable solid and (B) stable hydrated solid.

Figure 3.6. UV-Vis absorption spectra of CTAB in water.

X-ray diffraction (XRD) analysis was performed to study the crystalline nature of CTAB solution. Fig 3.7 shows the XRD pattern of CTAB solution, proving its highly crystalline nature.

Figure 3.7. XRD spectra of CTAB.

Fig 3.8 shows the transmitted power variation with a constant flow rate of 80 ml/hr. It was observed that the power increases to a maximum value and after few seconds it decreases exponentially. As the presence of the aqueous solution of CTAB in the flow cell, kept between crossed polarizers causes an increase in the transmitted power, it is the phenomenon of birefringence that plays a dominant role [93]. The CTAB flow cell can be regarded as a birefringent medium with birefringence $\Delta n = n_e - n_o$, where n_e and n_o are the refractive indices of extraordinary and ordinary rays that propagate with orthogonal polarizations, respectively.

Figure 3.8. Variation of output laser power with time at a constant flow rate of 80 ml/hr (0.5 M CTAB).

The change in the refractive indices will create a phase difference δ between the two rays, as it is emerged from the CTAB flow cell of depth, d [Equation (3.1)].

$$\delta = \frac{2\pi\Delta nd}{\lambda} \quad (3.1)$$

Ordinary index of refraction (n_o) is independent of the direction, while extraordinary index of refraction (n_e) depends on the orientation of the molecules. When the orientation of molecules varies, n_e also changes known as effective refractive index, n_{eff} and is given in equation (3.2).

$$n_{eff} = \frac{n_e n_o}{\sqrt{(n_e^2 \cos^2 \theta + n_o^2 \sin^2 \theta)}} \quad (3.2)$$

Therefore, the birefringence is modified as given in equation (3.3), where θ is the angle between direction of propagation and optic axis of the system.

$$\Delta n = n_e(\theta) - n_o \quad (3.3)$$

The transmitted light intensity emerging from the analyzer can be obtained using Jones calculus as given in equation (3.4) and more details regarding the matrix formulations are given in Appendix A (section A.2).

$$\begin{bmatrix} E_x' \\ E_y' \end{bmatrix} = J_A J_M J_P \begin{bmatrix} E_x \\ E_y \end{bmatrix} \quad (3.4)$$

Where E_x, E_y, E_x' and E_y' are the amplitudes of the incident and transmitted optical beam along the X and Y-axes, respectively. Here, J_A, J_M and J_P are the Jones vector for the analyzer, the birefringent medium (CTAB flow cell) and the polarizer, respectively.

$$\begin{bmatrix} E_x' \\ E_y' \end{bmatrix} = \begin{bmatrix} 1 & 0 \\ 0 & 0 \end{bmatrix} \begin{bmatrix} e^{i\delta_x} \cos^2 \psi + e^{i\delta_y} \sin^2 \psi & (e^{i\delta_x} - e^{i\delta_y}) \sin \psi \cos \psi \\ (e^{i\delta_x} - e^{i\delta_y}) \sin \psi \cos \psi & e^{i\delta_x} \cos^2 \psi + e^{i\delta_y} \sin^2 \psi \end{bmatrix} \begin{bmatrix} 0 & 0 \\ 0 & 1 \end{bmatrix} \begin{bmatrix} E_x \\ E_y \end{bmatrix} \quad (3.5)$$

Where δ_x and δ_y are the phase differences along horizontal (X), vertical (Y) axis, ψ is the angle between the optic axis and X-axis, respectively. The intensity of the light emerging from the analyzer can be obtained from Jones matrix and is given below.

$$I_t = \sin^2 \left(\frac{\pi(n_{eff} - n_o)d}{\lambda} \right) \sin^2(2\psi) I_0 \quad (3.6)$$

Where I_t , I_0 are the transmitted and input intensity, respectively. For particular flow rate, the birefringence is varied which affects the light intensity. Therefore, the change in the transmitted light intensity or power can be attributed to the birefringence of the medium. The exponential decrease in power after the initial increase was due to the absence of hydrated crystals in the transmission path. Since the observed variation is due to the presence of hydrated crystals, time for which the solution is pumped into the flow cell can also affect the transmitted intensity. Therefore the experiment is done for different time period of pumping. Fig 3.9 depicts the effect of pumping period at which the CTAB solution was injected into the flow cell.

Figure 3.9. Variation in output power for different pumping time. (a) Flow rate of 80 ml/hr was kept constant for 250 sec and (b) Flow rate of 80 ml/hr, switched off after 30 seconds.

The decay was fitted with a single exponential function and obtained the time constants. The exponential decrease in power seems to be faster (time constant, $t_1 = 101$ sec) when the flow rate was switched off after 30 sec than in the case of constant flow rate up to 250 sec (time constant, $t_2 = 299$ sec). This phenomenon is due to the settling of particles in the syringe.

The concentration of lyotropic liquid crystals also plays an important role in the crystalline nature of the material. We have also studied the effect of concentration in the transmitted intensity. Fig 3.10 demonstrates the variation of transmitted intensity for three concentrations of CTAB, 0.4M, 0.5M and 0.6M. As the concentration increases, the exponential decrease in intensity seems to increase.

Figure 3.10. Variation in transmitted power with respect to time for, 0.4M, 0.5M and 0.6M CTAB.

In order to avoid the sedimentation of particles in syringe, the CTAB solution was stirred continuously using a magnetic stirrer [Fig 3.4]. With this modified experimental setup the effect of flow rate (80 ml/hr) on the output laser intensity was monitored for about 200 seconds as shown in Fig 3.11. It was found that the output intensity remains constant after the initial increase, till the flow was switched off.

Figure 3.11. The variation of laser power with time at a flow rate of 80 ml/hr.

The effect of different flow rates on the output power was also monitored. At very low rates, there was no significant change in the intensity. For higher flow rates, intensity fluctuations were large. Fig 3.12 represents the variation of transmitted intensity for four different flow rates 40 ml/hr, 60 ml/hr, 80 ml/hr and 100 ml/hr. In this research, the source and detector were placed in the transmission path and the power increases in the presence of flow. Therefore, the phenomenon of scattering will have minimal effect on the transmitted power variation.

Figure 3.12. Variation of power with time for different flow rates.

However, in order to confirm the above hypothesis, the effect of scattering was analysed using integrated sphere technique. An integrating sphere can be used to measure either specular or diffuse reflection and provide a 180° field of view in the detection. Diffuse reflection is caused by multiple scattering from a rough surface or a layer nearby the surface. An interrogation area of about 1cm^2 of the flow cell was analysed and the percentage reflectance for three different experimental conditions was done. Reflectance percentage from the flow cell in the presence of CTAB is found to be comparatively low when compared with the reflectance from the flow cell alone. Thus, the scattering from the hydrated crystals in the flow cell was monitored to be significantly low and has found to be negligible. The percentage reflectance from one face of the flow cell as a function of wavelength is shown in the Fig 3.13.

Figure 3.13. Variation of percentage reflectance with wavelength.

3.4. Conclusion

In this chapter the effect of a lyotropic liquid crystalline material, cetyltrimethylammonium chloride (CTAB) in changing the polarization of the transmitted optical beam was investigated. Here the controlling parameter was the flow rate at which the fluid was injected into the channel. It was inferred that the lamellar structures formed in the surfactant below Krafft temperature are birefringent. Also the birefringence can be tuned by the flow rate of the liquid. The effect of absorption and scattering on the transmitted power is also considered. These results can be further incorporated and explored for the fabrication of optical components such as tunable polarizer, variable attenuator. Another category of liquid crystals,

thermotropic liquid crystal has been analysed on the same platform in the next chapter.

Chapter 4

Investigations into the Fluid-light Interactions in Non-Aligned Thermotropic Liquid Crystal

The propagation of laser beam in a non-aligned thermotropic liquid crystal and various fluid-light interactions are studied in this chapter. The effect of polarization and scattering on the transmitted power through the liquid crystal under external perturbation, the fluid flow, is demonstrated. It is found that the flow rate has a significant role in the modulation of refractive index of the medium leading to scattering and change in polarization.

4.1. Introduction

Liquid crystalline materials are capable of flowing like fluids having a certain degree of order in the arrangement of molecules. Since their discovery in 1980 by Reinitzer and Lehmann, thousands of compounds have been identified to show this peculiar phase of matter. As discussed earlier, liquid crystals possess a high degree of shape anisotropy. This results in dielectric and optical anisotropy, which is one of the key features of liquid crystals. The shape of the molecules in a thermotropic liquid crystal can be disc, or rod like and the phase transitions take place due to the variations in temperature [98]. The available forms of thermotropic liquid crystals can be classified as nematic, smectic and cholesteric phases depending upon the structure of the molecules or building blocks. In nematic liquid crystal (NLC), molecules have a long-range orientational order and will be in the fluid form whereas smectic phase possesses both positional and orientational order. The average orientations of molecules follow a helical path in cholesteric phase. Among different types of liquid crystals, nematics are the widely studied one. Also the simplest phase of liquid crystal is the nematic [10, 99].

A nematic liquid crystal can be considered as a medium in the fluidic state, with majority of the molecules oriented along a common direction defined by means of a unit vector $\mathbf{n}$, known as the director. Most of the liquid crystal molecules possess permanent dipole moments. Since $\mathbf{n}$ and $-\mathbf{n}$ are indistinguishable, molecules are packed in such a way that the net dipole moment is zero. Nematic liquid crystal is a

highly birefringent material, in which the refractive index can be tuned extensively with $\Delta n = 0.05$ and has a high orientational order [100]. Generally a nematic liquid crystal medium is considered to be uniaxial, characterized by the tensor order parameter given as

$$Q_{\alpha\beta} = s(n_{\alpha}n_{\beta} - \frac{1}{3}\delta_{\alpha\beta}) \quad (4.1)$$

The order parameter $Q_{\alpha\beta}$, can vary slightly from point to point depending on the constraints such as boundary conditions imposed by the walls and external perturbations. Depending upon the direction of the external perturbation, nematic liquid crystal undergoes deformation similar to a solid material.

4.1.1. Hydrodynamics of Fluids

Rheological studies of anisotropic fluids are different from that of ordinary isotropic fluids. First, let us consider the flow of isotropic fluids and analyze how it is different from anisotropic materials. The parameters needed for the description of fluid dynamics are position vector (r), velocity (v), density (ρ), pressure (p) and the forces acting on it. Consider the movement of a volume element, $dv = dxdydz$ of an isotropic fluid in space with respect to an applied body force f .

Figure 4.1. Representation of movement of fluid volume element at a velocity, v [10].

The equation of continuity can be written as

$$\nabla \cdot (\rho \vec{v}) = -\frac{\partial \rho}{\partial t} \quad (4.2)$$

Since the fluid medium is assumed to be incompressible

$$\rho(r, t) = \text{const} \quad (4.3)$$

The density is considered to be a constant and therefore equation (4.2) becomes

$$\nabla \cdot \vec{v} = 0 \quad (4.4)$$

Newton's second law describing the rate of acceleration of the fluid volume element is

$$\rho \frac{d\vec{v}}{dt} = \vec{f} \quad (4.5)$$

Where f is the applied body force. Equation (4.5) can be modified considering all the forces exerted on the fluid which includes the pressure gradient $(-\Delta p)$, internal viscous forces (f_{vis}) and externally applied force (F_{ext}) .

$$\rho \frac{dv}{dt} = -\Delta p + f_{vis} + F_{ext}$$

$$\frac{dv}{dt} = \frac{-\Delta p}{\rho} + \frac{\eta \nabla^2 v}{\rho} + \frac{F_{ext}}{\rho} \quad (4.6)$$

Equation (4.6) is the well known Navier-Stokes equation for incompressible isotropic fluid and the second term on the right-hand side of equation (4.6) represents the stress tensor (σ) [10].

The foremost characteristic of an anisotropic fluid is that it is composed of rod like molecules which are approximately aligned along a common direction. Thus, a translational motion of the fluid will result in an orientational motion and the dynamics is more complex than the isotropic fluids. The continuum theory of nematic liquid crystals (NLC), an anisotropic fluid was first formulated by Oseen and Zocher in 1920s and later it was re-examined by Frank [101, 102]. Later the constitutive equations explaining the mechanical behaviour of nematic liquid crystal were presented by Leslie and Ericksen. This the most widely used theory to describe the dynamics of nematic state [103-105]. According to Leslie-Ericksen theory, stress tensor also depends on the orientation and rotation of the molecules other than the velocity gradient.

In the absence of an external force, the NLC molecular alignment is such that the system has minimum free energy density, F_d . When an external force (F_{ext}) is applied, the molecular orientation changes and thus increases the free energy as given in equation 4.7 [100]. The increase in free energy can be expressed in terms of elastic constants, K_{11} , K_{22} , K_{33} , representing three different deformations like splay, twist and bend, respectively [Fig 4.2].

Figure 4.2. Various deformations of nematic liquid molecules (a) splay (b) twist (c) bend [10].

$$2F_d = K_{11}(\nabla \cdot n)^2 + K_{22}(n \cdot \nabla \times n)^2 + K_{33}|n \times \nabla \times n|^2 \quad (4.7)$$

By assuming the nematic as incompressible, the balance equations for mass, linear momentum and director (angular momentum) are given by

$$\nabla \cdot v = 0 \quad (4.8)$$

$$\rho \frac{\partial v}{\partial t} = f + \nabla \cdot \sigma \quad (4.9)$$

$$\rho_I \ddot{n} = G + g + \nabla \cdot \xi \quad (4.10)$$

Where ρ , v , f , σ , ρ_I , G , g and ξ are density, velocity, body force per unit volume, total stress, moment of inertia per unit volume, external director body force, intrinsic director body force and director stress tensor, respectively [106]. As discussed earlier, external forces or fields acting on NLC can cause translational as well as the orientational order of the director. The director body force and director stress tensor are the torque needed for changing the orientation (rotation) of the molecules and the corresponding stress on the director due to that force.

In this theory, the total stress (σ) and intrinsic director body force (g) are given in terms of angular velocity of the director, relative to that of the fluid (N) and the rate of the deformation tensor (A). Therefore, linear constitutive equations for σ , g and ξ can be written as follows [106].

$$\sigma = -pI - \frac{\partial F_d}{\partial \nabla n} \cdot \nabla n^T + \alpha_1 (nn \cdot A) nn + \alpha_2 nN + \alpha_3 Nn + \alpha_4 A + \alpha_5 nn \cdot A + \alpha_6 A \cdot nn \quad (4.11)$$

$$g = an - \beta \cdot \nabla n - \frac{\partial F_d}{\partial n} - \gamma_1 N - \gamma_2 n \cdot A \quad (4.12)$$

$$\xi = \beta n + \frac{\partial F_d}{\partial \nabla n} \quad (4.13)$$

Where p is the pressure, I is the unit tensor, γ_1 is the rotational viscosity, γ_2 is the irrotational torque coefficient, β is the Lagrange multiplier vector and $\alpha_1, \alpha_2 \dots \alpha_6$ are the six Leslie viscosity coefficients.

$$2A = \nabla_v + \nabla_v^T \quad (4.14)$$

$$2W = \nabla_v - \nabla_v^T \quad (4.15)$$

(Where, W is the rate of spin tensor)

$$\gamma_1 = \alpha_3 - \alpha_2 \quad (4.16)$$

$$\gamma_2 = \alpha_6 - \alpha_5 = \alpha_3 + \alpha_2 \quad (4.17)$$

$$N = \dot{n} - W.n \quad (4.18)$$

Flow induced molecular orientation take place when the total viscous torque density (Γ_{vis}) vanishes. The first term represents the effect of transient rotation and second term is the effect of deformation [10, 106].

$$\Gamma_{vis} = n \times [\gamma_1 N + \gamma_2 A n] \quad (4.19)$$

As pointed out earlier, in this chapter, Poiseuille flow of non-aligned liquid crystal molecules through a rectangular channel is analysed. Therefore, the velocity distribution follows a parabolic profile as given in equation (4.20) [107].

$$u_x = -\frac{1}{2\eta} f z(z-d) \quad (4.20)$$

Where η is the viscosity and f is the applied body force and d is the depth of the rectangular channel. From the above equation (4.20), the volumetric flow rate can be written as,

$$Q = \int_0^d u_x dy = \frac{1}{12\eta} f d^3 \quad (4.21)$$

In the case of a rectangular channel, shear rate and flow rate is related as given in equation (4.22) [108].

$$\dot{\gamma} = \frac{6Q}{wd^2} \quad (4.22)$$

Where, $\dot{\gamma}$ is the shear rate, Q is the flow rate, w and d is the width and depth of the channel.

There has been reports based on the director reorientation in nematic liquid crystals by the application of voltage, oscillatory shear and using Poiseuille flow [109-114]. In this chapter, the effect of Poiseuille flow, in a non-aligned nematic liquid crystal in altering the optical functionalities were studied and discussed in detail. Here, the NLC was introduced into the channel without any surface alignment. Therefore, the NLC molecules exist in multi-domains inside the flow cell. When an optical beam is transmitted through a non-aligned nematic liquid crystal cell, fluid-light interactions such as state of polarization variation, scattering and absorption can occur.

4.2. Experiment

4.2.1. Fabrication of the Flow Cell

A 25 mm long, 0.5 mm depth and 1 mm width rectangular flow cell was fabricated manually using microscopic glass substrate. The method of cell fabrication is similar to the one described in chapter 3. Flow cell was connected to a syringe

pump using silicone tubing with an outer diameter of 0.3 cm and inner diameter of 0.2 cm. The channel was cleaned before introducing the liquid crystal.

4.2.2. Properties of the Nematic Liquid Crystal

Nematic liquid crystal, 4-pentyl-4'-cyanobiphenyl (5CB, Merck Pte Ltd) was used for the analysis, at room temperature. The liquid crystal belongs to the cyanobiphenyl family. The chemical structure of 5CB is shown in Fig 4.3. Depending upon the length of the aliphatic chain, NLC can exhibit different phase behaviour. It remains in the nematic phase within a temperature range of 22 °C to 34 °C [72].

Figure 4.3.Chemical structure of 4-pentyl-4'-cyanobiphenyl (5CB) [115].

4.2.3. Experimental set up and Procedure

In order to analyze the change in the polarization under different flow rates, the flow cell was kept between crossed polarizers as shown in Fig 4.4. A light source of He- Ne laser of wavelength 632 nm was used for the investigation. A power meter (Thorlabs, Inc. PM100), interfaced to a computer was used to detect the power variation. The flow cell was kept in such a way that, the flow direction (X-axis) makes an angle of 90° with the polarization axis of the polarizer (Y-axis). The NLC was injected into the flow cell using a syringe pump (Cole Parmer, EW-74900-05). When the medium became stable, a particular flow rate was given for 50 seconds and

the output power was monitored. After switching off the flow, the transmitted power was monitored for 250 seconds. This was repeated for both low and high flow rates. Experiments were also performed by replacing the photodiode with a CCD camera (Pixelink-BL471U) to analyze the effect of scattering. The absorption spectrum was measured on a Shimadzu Corp. (UV-1601PC) spectrophotometer to monitor the contribution of absorption in the visible wavelength.

Figure 4.4. Schematic of experimental set up.

4.3. Results and Discussion

The UV-Vis absorption spectrum of the liquid crystal is shown in Fig 4.5. The spectrum shows that there is no absorption in the visible wavelength regime. Therefore, the effect of absorption in the power attenuation can be neglected. Other than absorption, there can be fluid-light interactions such as polarization and scattering that can contribute to the transmitted power variation.

Figure 4.5. UV- Vis absorption spectrum of 4-pentyl-4'-cyanobiphenyl.

The effect of polarization was analysed by monitoring the transmitted light intensity through the NLC flow cell kept between crossed polarizers. Fig 4.6 and Fig 4.7 shows the variation of transmitted power with respect to time for different flow rates. The experiments were repeated for a number of times (minimum 5 times each) and obtained results are shown in Fig 4.6 and 4.7 (more details, detailing the standard deviation and mean are given in Appendix D). The respective change in the transmitted power is attributed to the refractive index modulation of the medium due to the external perturbation (flow) [Eq (3.6)]. Upon the application of an external force, the shear stress on the NLC given as stress tensor ' σ ' [Equation (4.11)], will tend to change the director orientation. The reorientation of director in turn changes the phase difference and hence will change the transmitted intensity. The Jones matrix evaluation is similar to equation (3.4) in chapter 3 and more details are given in Appendix A (section A.2).

Figure 4.6. Variation of transmittance with respect to time for low flow rates, $Q = 1$ ml/hr to $Q = 5$ ml/hr.

Figure 4.7. Variation of transmittance with respect to time for high flow rates, $Q = 35$ ml/hr to $Q = 50$ ml/hr.

The experiment was also repeated with another wavelength of 514 nm (Argon-ion laser). Fig 4.8 illustrates the variation of transmitted intensity with respect to flow rate ranging from $Q = 1$ ml/hr to $Q = 50$ ml/hr. The experimental procedure adopted was the same as detailed in the earlier case.

Figure 4.8. Variation of transmittance for flow rates (a) $Q = 1$ ml/hr and $Q = 5$ ml/hr (b) $Q = 35$ ml/hr to $Q = 50$ ml/hr.

Fig 4.9 shows the CCD images of the transmitted laser beam for various flow conditions. At flow rate, $Q = 1$ ml/hr after 50 sec, the beam was expanded in the vertical direction as it was transmitted through the NLC flow cell [Fig 4.9 (a)]. After switching off the flow, within 200 sec [Fig 4.9 (b) and (c)], the laser beam expansion was reduced. This effect was more pronounced for higher flow rate, $Q = 50$ ml/hr [Fig 4.9 (d), (e) and (f)]. This can be attributed to the phenomenon known as beam fanning, a scattering effect.

Figure 4.9. CCD images of the flow cell at different flow conditions (a) $Q = 1\text{ ml/hr}$, $t = 50\text{ sec}$ (b) $Q = 0\text{ ml/hr}$, $t = 100\text{ sec}$ (c) $Q = 0\text{ ml/hr}$, $t = 200\text{ sec}$ (d) $Q = 50\text{ ml/hr}$, $t = 250\text{ sec}$ (e) $Q = 0\text{ ml/hr}$, $t = 350\text{ sec}$ (f) $Q = 0\text{ ml/hr}$, $t = 450\text{ sec}$.

Beam fanning is a nonlinear phenomenon, occurring due to scattering of light within the medium. There are reports on the beam fanning effect of many materials such as barium titanate and lithium niobate [116]. When a single beam is incident on a photorefractive material, the incident energy is transferred into scattered beam and is amplified, resulting in beam fanning. A non-uniform illumination of the material can also lead to beam expansion or beam fanning. This arises due to refractive index inhomogeneity [117]. Without any preferred orientation, liquid crystal molecules exist in multi-domains and the refractive index discontinuity at these domain boundaries is expected to cause the scattering phenomenon [72]. When a shear force (flow) is introduced into the material, refractive index modulation became more pronounced resulting in the transmitted beam expansion. To the best of our

knowledge, the demonstration of this phenomenon, in a multi domain nematic liquid crystal has not been reported. When the flow is switched on, beam was fanned out and this expansion increases with the flow rate. In the presence of a flow, change in the alignment of liquid crystalline molecules results in large refractive index modulation. Fig 4.6, Fig 4.7 and Fig 4.9 shows that there is a flow induced refractive index modulation taking place which results both in SOP change and scattering.

For the further analysis of the transmitted laser beam performances, images shown in Fig 4.9 (a) and (d) are converted to contour plots as shown in Fig 4.10 (a) and (b), respectively.

Figure 4.10. Image contours of laser beam transmitted through the flow cell under flow rate (a) $Q = 1$ ml/hr (b) $Q = 50$ ml/hr.

With this we can analyze the intensity distribution of the expanded beam. It can be inferred from Fig 4.10 (a) and (b) that the expansion of the beam along the major axis, at a flow rate of 50 ml/hr is higher compared to 1 ml/hr.

4.4. Conclusion

In summary, it can be concluded that an external force applied on the NLC, results in a refractive index modulation of the medium. This again causes SOP variation and scattering. Here the NLC molecules are non-aligned and the scattering results due to the existence of multi-domains of molecules. Therefore, it is expected to reduce the scattering by making the molecular arrangement homogenous and also reducing the flow cell dimension. This has been demonstrated in the next chapter.

Chapter 5

Poiseuille Flow of Homeotropically and Planar Aligned Nematic Liquid Crystal

The response of homogeneous or aligned nematic liquid crystal molecules with respect to an external perturbation is analysed. Furthermore the concept of liquid crystal based tunable optofluidic polarizer is proposed and illustrated. The tunability in the polarization is achieved by allowing the liquid crystal to flow through a rectangular channel at a suitable flow rate for a particular period of time. It is demonstrated that pressure driven flow perpendicular to the director $\mathbf{n}$, can induce a reconstruction of the molecular alignment from vertical to horizontal state through several intermediate states. Hence the polarization state of the transmitted beam can be changed. Further, the topological changes in nematic liquid crystal molecules under Poiseuille flow are analysed in detail, employing conoscopy and optical transmittance studies. The behaviour of homeotropically and planar aligned nematic liquid crystal molecules in response to flow are investigated.

5.1. Introduction

In most liquid crystal applications, the confinement of liquid crystal molecules between the boundary surfaces is very important. Without any preferred orientation, the NLC medium exists in multi-domains with different alignment in each of the domain [72]. Thus, it appears as a highly viscous fluid resulting in large scattering due to the refractive index discontinuity at these domain boundaries. Therefore, to reduce the effect of scattering, suitable boundary conditions are applied to the cell substrates to make the NLC medium homogeneous. As the scattering coefficient is inversely proportional to the cell thickness, reducing the distance between the confining surfaces can also significantly decrease the amount of scattering [10].

Generally there are two types of liquid crystal orientation; planar and non planar textural arrangements. There are different methods for attaining the prerequisite molecular arrangement. All the techniques strongly depend upon certain parameters such as dimension of the confining medium (cell thickness), elastic properties, surface boundary conditions and external forces. The ability of liquid crystal molecules to align along a preferred direction is an important aspect in most of the applications. Two basic alignments, namely, homeotropic and planar can be accomplished in different ways as shown in Fig 5.1. In homeotropic anchoring, the molecules tend to orient perpendicular to the substrate surface whereas the molecules have parallel orientation in planar alignment. Different types of methods to achieve

the desired orientation are available such as rubbing, fabrication of microgrooves, photoalignment, oblique evaporation, and chemical surface treatment.

Figure 5.1. Schematic representation of (a) planar and (b) homeotropic anchoring of NLC molecules [10].

The first section of this chapter, analyzes the effect of Poiseuille flow in homeotropically aligned NLC molecules. This is obtained by depositing a monolayer of silane compound on the substrates. The polar head group gets attached to the substrates and the hydrocarbon tails point away from the surface. The interaction between the hydrocarbon tails and liquid crystal molecules makes it to orient normal to the surface. The second section, describes the planar alignment of NLC molecules under Poiseuille flow. A cationic surfactant was used for providing planar alignment on the substrate surface. In both cases, the effect of flow on the molecular reconstruction was analysed. As the molecular reorientation take place, polarization of the transmitted beam also varies. In this context, a flow dependent phase shift with preferentially aligned nematic liquid crystal was proposed and illustrated.

The topological changes occurring in the flow cell, in the presence of pressure driven flow have been verified by employing interferometric method, conoscopy and

optical birefringence measurements. A brief description of different alignment techniques and the principle of conoscopy based evaluations are given in sections 5.1.1 and 5.1.2, respectively.

5.1.1. Different Alignment Techniques

Rubbing technology has been still the dominant method for the planar alignment of NLC molecules, first reported by Mauguin [118]. This is usually done by coating a polymer on the substrate surface and then rubbing it with a velvet cloth in the preferred direction. By rubbing, microgrooves are formed on the surface and the molecules get aligned along the grooves. Even though this method is used in many applications there are many disadvantages such as generation of dust, unevenness and less controllability. It is also possible to fabricate microgrooves on the surface using photolithographic technique and the alignment of NLC molecules is similar to rubbing method.

Another technique, photoalignment relies on the photoisomerization process, to control the NLC molecular alignment on the surface. To initiate the phenomenon of isomerism, NLC is doped with a dye, usually azobenzene. The major advantage is that, it is possible to change the alignment from planar to homeotropic by changing the properties of exposed light such as wavelength and polarization. But this method is not yet used for mass production due to the high cost.

In oblique evaporation method, a suitable oxide compound is coated on the substrate surface and the micro columnar structure formed on the evaporated film

aligns the liquid crystal molecules. The micro patterns formed on the film critically depends on the angle between the normal to the substrate surface and the direction of evaporation. Silicon monoxide (SiO) is usually used as the oxide compound since it is easily evaporated when heated. Any non-uniformity in the evaporated film thickness affects the liquid crystal molecular alignment and therefore this method is not suitable for many applications.

Liquid crystal alignment can also be done by applying chemical treatment using surface active agents. This method is generally used in two ways. Either with the treatment of substrate surface with suitable surface active compounds or by mixing surface active agents with liquid crystalline materials. For the latter case, surface active chemical is mixed with suitable solvent and then added to the liquid crystalline medium. The solvent is removed thereafter from the medium. In the former case, surface treatment is usually done in two ways, (i) dipping the substrates into solutions of surface active chemicals like silicon compounds, surfactants *etc*, (ii) exposing the surface to the vapour of suitable silane compounds.

The method adopted in this work is by dip coating with suitable compounds to form self assembled monolayers (SAM). Depending on the compounds, homeotropic or planar alignment of nematic liquid crystal molecules can be achieved. Self assembled monolayer was formed by the adsorption of surfactant molecule on to the substrate. This depends on the affinity of the head group of surfactant molecule for the substrate surface. The SAM technique has several advantages when compared to

other methods such as eliminating the need of rubbing and can provide planar or homeotropic alignment depending upon the concentration and type of the compound. Examples of two such chemical compounds are dimethyloctadecyl [3-(trimethoxysilyl) propyl] ammonium chloride (DMOAP) and cetyltrimethylammonium bromide (CTAB). Here SAM of the former one induce homeotropic alignment whereas the latter one provide either planar or homeotropic depending on the process and concentration [119].

5.1.2. Conoscopy: Interference in Nematic Liquid Crystal

Conoscopy is an optical interferometric technique, traditionally used to investigate the orientation of optic axis in anisotropic materials. This method is widely applied in areas such as material sciences, biological studies and mineralogy. The principle of conoscopy is based on the inherent birefringence of anisotropic materials. When a highly convergent beam is incident on such a medium, each ray at an angle with the optic axis effectively decomposes into two mutually orthogonal components. These components, upon exit from the medium create a phase difference and will result in interference fringes after recombination. Based on the interference pattern obtained, it is possible to verify the molecular orientation in the liquid crystal medium.

As stated above, nematic liquid crystal is an anisotropic material in which the optical properties depend on the direction of propagation and polarization of the light waves. When a plane polarized light with amplitude E is incident on an anisotropic

medium, at an angle θ with the optic axis, each ray is divided into two rays. These rays, *e* (extraordinary) and *o* (ordinary) have different indices of refraction, vibrating in mutually orthogonal directions D'' and D' respectively as shown in Fig 5.2.

Figure 5.2. Representation of vibration components transmitted by polarizer and analyzer [120].

After emerging from the flow cell, these rays create a phase difference δ between them and can bring to interfere if an analyzer is kept after the NLC cell. Transmitted intensity obtained from the Fig 5.2 is given in equation (5.1) [120].

$$I_t = E^2 [\cos^2 \chi - \sin 2\phi \sin 2(\phi - \chi) \sin^2(\delta/2)] \quad (5.1)$$

In the above relation, I_t is the resulting light intensity, E is the amplitude of the incident wave, χ is the angle between the polarization directions of the polarizer and analyzer and ϕ is the angle between D' and the polarization direction of polarizer.

When the analyzer and polarizer are kept crossed ($\chi = \pi/2$), equation (5.1) can be rewritten as

$$I_t = E^2 \left\{ \sin^2 2\phi \sin^2(\delta/2) \right\} \quad (5.2)$$

A highly convergent linearly polarized beam, when traversed through the NLC flow cell kept between crossed polarizers results in an interference pattern. The intensity at each point corresponds to a particular direction through the flow cell. Thus, from equation (5.2), it can be seen that the intensity depends on the values of ϕ and δ . The points where ϕ is a constant is known as isogyre and whereas those along δ is a constant are called isochromates. Therefore, the curves along $\sin 2\phi = 0$ and $\sin \delta/2 = 0$ are known as principal isogyre and principal isochromate. The position of isogyre mainly depends on the angle between the direction of vibration in the crystal and that of the vibration passed by the polarizer or analyzer. For isochromates, the intensity at each point depends on the angle of incidence [72].

5.2. Experiment

A diode laser of wavelength 405 nm was used as the light source and the conoscopic experimental set up is illustrated in Fig 5.3. The flow cell was kept between crossed polarizers and the resulting interference pattern was captured using a charge-coupled device (Pixelink-BL471U). The experimental setup for observing the flow dependent optical transmittance is similar to Fig 5.3, except that the flow cell

was placed in the path of a parallel optical beam. CCD was replaced by a power meter as the detector. Both homeotropically and planar aligned NLC flow cells were considered.

Figure 5.3. Experimental set up of optical conoscope.

5.2.1. Fabrication of the Flow Cell

The glass substrates used for the fabrication of channel were treated with octadecyltrimethoxysilane (OTMS, Aldrich Chem. Co.) to induce homeotropic alignment. A solution of isopropanol and OTMS in de-ionized water was prepared and sonicated for 5 minutes. After cleaning the glass substrates, it was vertically dipped into the solution and rapidly removed. The coated glass substrates were then heated in an oven at 120°C for 30 minutes. Thus, a monolayer of OTMS was deposited on the glass by dip-coating method.

For planar alignment, the glass substrates were treated with an aqueous solution of CTAB (Aldrich Chem. Co). A monolayer of CTAB was deposited on the glass by dip-coating method and allows it to dry. Here a low concentration of CTAB

(0.5M) was used for inducing planar alignment of molecules since higher concentration induces homeotropic alignment.

A flow cell of dimension, 2.5 cm (length) x 1 mm (width) x 50 μm (depth) was fabricated using microscopic glass slides of refractive index 1.52. The sides of the cell were defined by placing a strip (double sided tape) of 50 μm thick, between the upper and lower glass slides. A diagram demonstrating the method of cell fabrication is shown in Fig 5.4. The flow cell was filled with nematic liquid crystal (5CB) using a syringe pump and silicone tubing with an outer diameter of 0.3 cm and an inner diameter of 0.2 cm. The cell was kept undisturbed for some time to reach a steady state condition.

Figure 5.4. Schematic of the method of channel fabrication.

5.3. Results and Discussion

5.3.1. Topological Changes in a Homeotropically Aligned NLC Cell

Consider a homeotropically aligned NLC flow cell sandwiched between a pair of crossed polarizers. In the static case, NLC molecules were oriented perpendicular to the cell wall (along the Z-axis), due to the surface anchoring. When an external force is applied, normal to homeotropically aligned nematic sample, a combination of splay and bend deformations occurs [10]. The director, $\mathbf{n}$ can be represented as $\mathbf{n} = (\sin \theta, 0, \cos \theta)$, assuming the twist angle to be zero. The boundary conditions imposed by the cell wall, causes the molecules in the vicinity and away from it, to align differently with respect to flow.

$$2F_d = K_{11}(\nabla \cdot \mathbf{n})^2 + K_{33}|\mathbf{n} \times \nabla \times \mathbf{n}|^2 + F_{ext} \quad (5.3)$$

Thus when the flow is switched on, inside the channel there are two alignment regions. Near the walls there is a transition layer of thickness $e_1 = \sqrt{Kwd^2/6\eta Q}$ at

which the molecules tend to align with the flow at an angle θ_L (Leslie angle). In the central region of the channel (where $\theta_L \cong 0$), the layer thickness is given as

$e_2 = d^{1/3}e_1^{2/3}$, K being the elastic coefficient, η is the viscosity, d is the distance between cell walls and Q , the flow rate [100]. The flow induced molecular alignment

near (region I) and away (region II), from the boundary surface is illustrated in Fig 5.5.

Figure 5.5. Schematic representation of Poiseuille flow in a rectangular channel [100].

Therefore, at high flow rates, in the middle of the cell, the molecules are preferred to be aligned along the direction of flow. Thus in the presence of flow, a switching between two topological textures take place. Fig 5.6 demonstrates the alignment of NLC molecules in the cell and how the orientation of molecules is affected by the flow, along the channel depth.

Figure 5.6. (a) Schematic representation of liquid crystal molecular arrangement inside the channel (b) Different nematic textures in the stable vertical state (V) and under flow horizontal state (H).

The molecular rearrangement with respect to flow and corresponding SOP variation was monitored by employing the methods of conoscopy and optical transmittance studies. The following sections illustrate the principle of these two methods.

5.3.1.1. Determination of Phase Retardation at Oblique Incidence Using Convergent Beam (Conoscopy)

The use of convergent beam has two consequences: 1) the optical path of each ray is different 2) the directions of vibrations corresponding to each of the ray are different. As the rays became more inclined with respect to the surface normal, path difference increases proportionally and thus varying the phase difference. Each degree of inclination is considered as an optical cone. The birefringence (Δn) will be different for each of the cone of rays. Also as the degree of inclination changes, the direction of vibration of fast and slow rays varies. Therefore, the rays with direction of vibration that coincide with that of polarizer or analyzer will appear as dark due to extinction.

Referring to Fig 5.7 (a), consider a beam of light AB incident on the surface at an angle θ with respect to the optic axis. The optic axis considered to be along Z-axis. Due to double refraction, it splits into two components making angles θ_e and θ_o with respect to the optic axis.

Figure 5.7. (a) Schematic of the trajectory of rays at oblique incidence (b) Double refraction in a uniaxial medium with optic axis perpendicular to the boundary [72].

From the Fig 5.7 (a), phase difference can be written as [72]

$$\delta = n_e AB + BD - n_o AC \quad (5.3)$$

Applying Snell's law of refraction and simplifying equation becomes

$$\delta = \frac{2\pi}{\lambda} [n_e \cos \theta_e - n_o \cos \theta_o] d \quad (5.4)$$

For light incident at an angle with respect to the optic axis (oblique incidence), the phase difference is slightly modified as shown in equation (5.4). Where n_e (slow axis), n_o (fast axis) are the extraordinary and ordinary refractive indices of the two rays, respectively and d is the depth of the flow cell. Fig 5.7(b) demonstrates the double refraction occurring at the boundary, consisting of two parts. The wave number k_1 corresponding to the direction of propagation of ordinary wave follows a

sphere. The other, k_2 corresponding to the extraordinary wave follows an ellipsoid of revolution and thus depends on the direction of propagation.

Therefore, a highly convergent beam traversed through the NLC flow cell, kept between crossed polarizers, results in interference fringes which consist of melatope, isogyre and isochrome as shown in Fig 5.8. In the static case, NLC molecules were oriented perpendicular to the substrate surface due to surface anchoring. The centre of the interference figure, known as the melatope, corresponds to the rays travelling parallel to the optic axis with zero-order retardation. As discussed earlier, for the rays with vibrations parallel to that of polarizer ($\phi = 0$), will get extinct after the analyzer and results in principal isogyres. The principle isochrome corresponds to rays with phase shift, $\delta = 2\pi$.

Figure 5.8. Schematic of convergent light propagation through the NLC flow cell.

Fig 5.9(a-l) shows the recorded interference pattern obtained when a convergent beam is transmitted through the NLC flow cell for different flow conditions. In the stable state, a dark cross superimposed with a circular fringe was

obtained [Fig. 5.9(a)]. Once the flow, $Q \perp n$ is switched on, the change in the fringe pattern [Fig. 5.9(b-l)] can be observed due to the fluctuation in the molecular arrangement. Isochrome evolves from circle to hyperbola; isogyre get splits and gradually became two symmetrical hyperbolas which escape from the field of view [Fig. 5.9(b)-(d)].

For crystals with uniaxial symmetry, change in the shape of isochrome occurs when the angle between optic axis (OA) and optical beam path varies. This is well documented in the literature [120]. There will be not any change in isogyre and will remain crossed. The splitting of isogyre is a characteristic feature of biaxial crystals and therefore it is presumed that a transient biaxiality was exhibited by the medium during the reconstruction of NLC molecules from vertical (V) to horizontal (H) state.

Figure 5.9. Recorded CCD images of the flow cell at a flow rate, $Q = 20 \mu\text{l/hr}$ for a particular period of time.

Nematic liquid crystals normally consist of rod shaped molecules with cylindrical symmetry. The NLC used in this investigation, is considered to be uniaxial with symmetry group denoted by the symbol, $D_{\infty h}$ (Schoenflies notation) [100]. However, there can be chances of deviation from cylindrical symmetry leading to molecular biaxiality. Here the term biaxial indicates the presence of two axes along which a plane polarized optical beam can travel without any change in the state of polarization. In 1970, Freiser had predicted the existence of biaxial nematic state based on the generalized Maier–Saupe theory [121]. According to this theory, a reduction in the molecular symmetry could lead to the formation of a biaxial nematic phase. After Freiser, the existence of biaxial nematic phase was reported ten years later, by Yu and Saupe, for a ternary lyotropic system (potassium laurate-1-decanol- D_2O) for a particular temperature and concentration range [122].

The basic difference of biaxial phase from the commonly found uniaxial is that they exhibit additional orientational order along other two axes, $\mathbf{m}$ and $\mathbf{l}$. They are known as secondary directors. Therefore, in a biaxial medium, an optical wave experience three different velocities along these directions [123]. Schematic representations of uniaxial and biaxial nematic liquid crystal molecules are shown in Fig 5.10.

Figure 5.10. Representation of uniaxial and biaxial nematic liquid crystal molecules [123].

Biaxiality can be induced by various factors such as temperature, bending of molecules, addition of dopants, and presence of defects. First thermotropic biaxial state was found by Malthête *et al.* in 1986 [124]. Optical methods and X-ray diffraction studies were done to prove the biaxial behaviour. Biaxial nematic phase was also reported by the mixing of rod and disc shaped molecules [125, 126].

In recent years, it has been demonstrated that the application of strong external forces such as electric field and mechanical stress can induce biaxiality in otherwise uniaxial medium [127-129]. In uniaxial nematic, orientational order along the long molecular axis, $\mathbf{n}$ is more prevalent and orientations along short axes are uncorrelated except at short range. An external perturbation applied perpendicular to the direction $\mathbf{n}$, breaks the rotational symmetry along the long molecular axis. This results in a preferential alignment for the transverse axes of the molecules. Therefore, the nematic tensor order parameter changes, resulting in eigen value exchange problem. This concept is well explained using Landau–de Gennes theory by many researchers [127, 128, 130]. As discussed above, the average direction of orientation

of molecules is characterized by a unit vector, director $\mathbf{n}$ (which is considered as the optic axis). The flow $\mathbf{Q} \perp \mathbf{n}$ causes a heavy distortion in the molecular arrangement and generates a transient biaxiality during the transition. The biaxiality generated in this way is quite different from the intrinsic biaxiality, first observed by Freiser [121].

Fig 5.9(g) shows a fuzzy black cross known as flash figure [131]. Uniaxial flash figure is formed when the optic axis is nearly parallel to either polarizer or analyzer. Fig 5.9(h) consist of a set of dark hyperbolic fringes (upper and lower) which is usually obtained when the optic axis lies parallel to the substrate surface (H state) [72]. As time proceeds in the presence of flow, molecules continue to change the alignment and then finally attains a saturation level at H state [Fig.5.9 (g)-(l)]. Therefore, flow induces a change in the molecular alignment from V to H state through several intermediate stages as shown by the recorded images. After switching off the flow, molecules tend to relax to the initial position (V state) gradually. At a very low flow rate, the molecular reconstruction is found to be very slow. Since the rotation of molecular orientation influences the optical polarization, the above mentioned reconstruction in the nematic texture results in a SOP variation. This is illustrated using experiments and theory in the next section.

5.3.1.2. Jones Matrix Evaluation of the NLC Flow Cell (Normal Incidence)

Light propagation through a birefringent medium consists of two normal modes with well defined phase velocities and polarization directions. The NLC flow cell was kept in the transmission path of parallel beams. The transmitted intensity

through the NLC flow cell kept between the crossed polarizers was monitored. An expression for transmitted intensity can be obtained from the evaluation of Jones matrix. Jones matrices of the polarizer, NLC medium and analyzer are given as J_P , J_A and J_M , respectively [72].

$$J_P = \begin{pmatrix} \cos^2 \alpha & \cos \alpha \sin \alpha \\ \sin \alpha \cos \alpha & \sin^2 \alpha \end{pmatrix} \quad (5.5)$$

$$J_A = \begin{pmatrix} \cos^2 \beta & \cos \beta \sin \beta \\ \sin \beta \cos \beta & \sin^2 \beta \end{pmatrix} \quad (5.6)$$

$$\begin{aligned} J_M &= \begin{pmatrix} e^{i\delta_x} \cos^2 \psi + e^{i\delta_y} \sin^2 \psi & (e^{i\delta_x} - e^{i\delta_y}) \sin \psi \cos \psi \\ (e^{i\delta_x} - e^{i\delta_y}) \sin \psi \cos \psi & e^{i\delta_x} \cos^2 \psi + e^{i\delta_y} \sin^2 \psi \end{pmatrix} \\ &= \begin{pmatrix} e^{-i\delta/2} \cos^2 \psi + e^{i\delta/2} \sin^2 \psi & -i \sin \delta/2 \sin 2\psi \\ -i \sin \delta/2 \sin 2\psi & e^{-i\delta/2} \sin^2 \psi + e^{i\delta/2} \cos^2 \psi \end{pmatrix} \end{aligned} \quad (5.7)$$

$$J = \begin{pmatrix} \cos^2 \beta & \cos \beta \sin \beta \\ \sin \beta \cos \beta & \sin^2 \beta \end{pmatrix} \begin{pmatrix} e^{-i\delta/2} \cos^2 \psi + e^{i\delta/2} \sin^2 \psi & -i \sin \delta/2 \sin 2\psi \\ -i \sin \delta/2 \sin 2\psi & e^{-i\delta/2} \sin^2 \psi + e^{i\delta/2} \cos^2 \psi \end{pmatrix} \begin{pmatrix} \cos^2 \alpha & \cos \alpha \sin \alpha \\ \sin \alpha \cos \alpha & \sin^2 \alpha \end{pmatrix} \quad (5.8)$$

Where, ψ is the angle between optic axis and X-axis. Substituting the values of α ,

β , ψ , Jones matrix can be written as

$$J = J_A J_M J_P = \frac{1}{2} \begin{pmatrix} 1 & -1 \\ -1 & 1 \end{pmatrix} \begin{pmatrix} e^{i\delta/2} & 0 \\ 0 & e^{-i\delta/2} \end{pmatrix} \begin{pmatrix} 1 & 1 \\ 1 & 1 \end{pmatrix} \quad (5.9)$$

More details regarding the concept and calculation of Jones matrix are given in Appendix A (section A.2.1). From Jones matrix, an expression for the transmitted intensity can be obtained and is given in equation (5.10). Therefore, transmitted intensity can be written as

$$I_t = 2 \sin^2 \frac{\delta}{2} \cos^2 2\psi \quad (5.10)$$

Maximum occurs at corresponding values of phase difference δ or phase retardation, $\Delta n.d$ (where $\Delta n = (n_e - n_o)$ is the LC birefringence and d, the cell depth) as given in equation 5.11.

$$\left. \begin{aligned} \delta &= \pi, 3\pi, 5\pi, \dots \\ \Delta n.d &= \frac{\lambda}{2}, \frac{3\lambda}{2}, \frac{5\lambda}{2}, \dots \end{aligned} \right\} \quad (5.11)$$

Fig 5.11 (a) illustrates the transmitted intensity variation at a flow rate of 20 $\mu\text{l/hr}$ with respect to time. The homeotropic NLC flow cell behaves like an isotropic medium, in the absence of flow due to zero birefringence ($n_e - n_o = 0$). As the flow is switched on, molecules start reorientation, resulting in the rise of transmitted intensity due to increase in the birefringence. Fluctuation in the intensity can be observed after the initial rise which is mainly due to the transient biaxiality.

Figure 5.11. (a) Variation of transmittance with respect to time at a flow rate, $Q = 20 \mu\text{l/hr}$. (b) Phase shift variation as a function of time for four different flow rates, $Q = 20, 40, 60$ and $80 \mu\text{l/hr}$.

In Fig 5.11(a), two peaks were observed at which the molecular alignment results in maximum birefringence (H state). After sometime, majority of the molecules gets reoriented by the flow so that the director, n (OA) became parallel to the flow direction. Increment in the flow rate, stimulate the rotation of NLC molecules and reaches the saturation position (H state) at a faster rate. As evident from Fig 5.11 (a) and (b), phase shift can be well controlled using the flow rate for a particular period of time. Thus it is possible to achieve the required phase shift by the Poiseuille flow of liquid crystal, which is a simple method to implement.

5.3.2. Flow Induced Transition in a Planar Aligned NLC Cell

The effect of flow in planar aligned NLC molecules, confined in a rectangular flow cell was monitored. Transmitted intensity was also analysed to verify the corresponding change in the polarization of the optical beam. Due to the anisotropic

nature of the nematic liquid crystal, transmission through the material placed between the crossed polarizers depends on the angle of incidence and the optic axis orientation. Here the optic axis of the planar aligned NLC medium is considered to be along the substrate surface, which is along the orientation of the director, $\mathbf{n}$. Fig 5.12 demonstrates the light propagation through the planar aligned NLC medium.

With reference to equation (5.2), maximum transmission is given by $\sin 2\phi = \pm 1$ which occurs for $\phi = \frac{\pi}{4}, \frac{3\pi}{4}, \frac{5\pi}{4}, \dots$. Minimum intensity occurs at points where $\sin 2\phi = 0$ or $\phi = \frac{\pi}{2}, \frac{3\pi}{2}, \frac{5\pi}{2}, \dots$. The emergent beam from the NLC cell will remain at the same polarization state, if the phase retardation is a multiple of 2π . Therefore, the optical beam gets extinct by the analyzer and remains dark. Thus the conoscopic pattern for parallel aligned NLC medium consists of a family of dark hyperbolic fringes known as isochromates.

Figure 5.12. Representation of planar aligned NLC molecules in the flow cell.

Fig 5.13(a)-(c) represents the conoscopic patterns obtained when a flow, Q is initiated parallel to the molecular orientation. Referring to Fig 5.13(a), in the steady state (in the absence of flow), interference pattern consists of a set of hyperbolic fringes as reported in literature [72].

Figure 5.13. Recorded CCD images of the flow cell at a flow rate, $Q = 20 \mu\text{l/hr}$ for a particular period of time.

As time proceeds [Fig 5.13(b) and (c)], it was observed that the upper and lower hyperbolas shift to the end slightly. This can be presumed due to a small tilt of the NLC molecules from the X-axis. Here the flow introduced is parallel to the molecular alignment, $Q \parallel n$. Therefore, it can be seen that there is no appreciable change in the conoscopic patterns.

For planar aligned NLC medium, the molecules are oriented at an angle $\theta = 90^\circ$ with respect to the Z-axis and $\psi = 0^\circ$ with respect to the X-axis in the stable state. This orientation causes the NLC medium to act as a retardation plate with maximum retardance for normally incident beam. Fig 5.14 illustrates the transmission through the NLC flow cell at different flow rates. The intensity remains

approximately the same in the presence of the external perturbation. Thus from Fig 5.13 and Fig 5.14, a flow introduced in the same direction as director, $\mathbf{n}$ cannot cause much change in the molecular orientation.

Figure 5.14. Variation of transmittance with respect to time for different flow rates.

Jones matrix evaluation of the planar aligned nematic flow cell kept between crossed polarizers was also done. Corresponding Jones matrices of the polarizer, NLC medium and analyzer are given as J_P , J_A and J_M , respectively and more details are given in Appendix A (Section A.2.2)

$$J = J_A J_M J_P = \frac{1}{2} \begin{pmatrix} 1 & -1 \\ -1 & 1 \end{pmatrix} \begin{pmatrix} e^{-i\delta/2} & -i\sin\delta/2 \\ -i\sin\delta/2 & e^{i\delta/2} \end{pmatrix} \begin{pmatrix} 1 & 1 \\ 1 & 1 \end{pmatrix} \quad (5.12)$$

Transmitted intensity is given as

$$I_t = 2 \sin^2 \delta / 2 \quad (5.13)$$

Maximum occurs at

$$\delta = \pi, 3\pi, 5\pi, \dots \quad (5.14)$$

5.4. Conclusion

A study on the flow induced reconstruction of homeotropically aligned nematic liquid crystal molecules was carried out using conoscopy and optical transmittance. The effect of flow in a planar aligned nematic liquid crystal medium was also demonstrated. It was inferred that, a force applied parallel to $\mathbf{n}$, has not caused any appreciable change in the molecular alignment. The concept and developed probe can find potential optics and lab-on-a-chip applications.

Chapter 6

Conclusion and Future Work

This chapter concludes the thesis by pointing out the salient and original contributions made during the course of PhD. The future research directions are identified and explained by highlighting the potential research challenges ahead.

6.1. Conclusion

In this chapter, the overall conclusions of the thesis are presented. In the thesis, dynamic properties of liquid crystalline materials have been investigated for the realization of optofluidic polarization devices. Flow induced variations in lyotropic and thermotropic liquid crystals were done both experimentally and theoretically.

6.1.1. Major contributions

The major contributions of this research thesis can be summarized as follows:

- ❖ Conceptualization and design of a liquid crystal based optofluidic polarizer for lab-on-a-chip applications.
- ❖ Fundamental research into the various fluid-light interactions in a flow driven liquid crystal medium.
- ❖ Lamellar phase observed in lyotropic material below Krafft temperature was analysed and the exhibited optical birefringence property was demonstrated experimentally.
- ❖ Illustration of SOP variation and beam fanning in a non-aligned thermotropic liquid crystalline material under Poiseuille flow.
- ❖ Demonstration of flow induced refractive index modulation in aligned thermotropic liquid crystals.
- ❖ Evaluation of transmittance of aligned and non-aligned NLC medium using Jones matrix.

A liquid crystal based optofluidic polarization device was proposed and illustrated in this thesis. Such a system can be integrated into a heterogeneous chip with built-in microfluidic delivery. In commercially available LC devices, electric field is used to control the device parameters. Even though the existing LC polarization devices have many merits compared to other techniques, the complex configuration causes a hindrance for the integration into on-chip devices. Therefore, a new way to tune the optical properties of LC materials was demonstrated in the thesis.

Among various techniques employed for refractive index modulation, the method of applying a shear force or flow for the same was the least explored one. Variation in the refractive index, in response to an external shear depends on the molecular structure of the medium. Liquid crystalline materials, was found to be an excellent choice for the same. In LC, as the translational and orientational motions are linked, a pressure difference across the medium can induce a flow and thus creates a change in the molecular alignment. A realignment of LC molecules varies the optic axis of the system and thus can change the refractive index (Δn). Using the aforementioned liquid crystal characteristics, it is possible to tune the optical polarization with the applied shear or flow. Compared to the existing techniques, flow induced SOP variation is a simple method to implement. Optical devices such as variable attenuator and phase retarder can also be designed and fabricated using the above mentioned method.

In order to understand the shear or flow induced effects in liquid crystals, both lyotropic and thermotropic liquid crystals were analysed. Analysis on the fluid-light interactions in lyotropic liquid crystal material (CTAB) was investigated in detail. The hydrated crystals formed in the LC material were monitored to exhibit birefringence and was characterized by using absorption spectroscopy, optical microscopy and X-ray diffraction studies. The contribution of absorption and scattering was found to be insignificant. Also it was noted that the observed birefringence can be controlled with respect to flow. Rather than altering the molecular order, it was the presence of hydrated crystals that caused the corresponding SOP variation. Therefore, to analyze the molecular reorientation with respect to flow, thermotropic liquid crystal was considered.

Nematic liquid crystal, a thermotropic liquid crystal (4-pentyl-4'-cyanobiphenyl) was used in a similar configuration. Experiments were carried out to study the molecular dynamics in a non-aligned NLC medium. In non-aligned NLC medium, molecules exist in multi-domains. Refractive index inhomogeneity near the boundaries of each domain resulted in both scattering and SOP variation. A phenomenon known as beam fanning, a scattering effect was observed and analysed in detail. Scattering losses, due to the existence of multi-domains can be reduced by providing a homogeneous alignment of NLC molecules. This was done by using surface treated flow cell and also by reducing the channel depth [10, 72]. Also, in the non-aligned state, molecular realignment takes place differently in different domains

in the presence of flow. Therefore, the response of aligned thermotropic liquid crystal with respect to pressure driven flow was demonstrated subsequently. The same thermotropic NLC, as specified above was used. Different types of alignments and the corresponding flow driven transitions were monitored. Homeotropically and planar aligned NLC molecules were obtained by providing suitable surface treatment on the cell substrates.

In the presence of flow, a complete reorientation of the molecules was monitored for homeotropically aligned NLC. The topological changes occurring across the nematic medium was analysed using the interferometric technique, conoscopy. The corresponding changes in the conoscopic patterns, demonstrated the NLC molecular reconstruction in the presence of flow. As stated above, the flow induced molecular reorientation varies the polarization of the transmitted beam. Therefore, the variation in the state of polarization was monitored from the transmitted intensity variations. Also a transient biaxiality was observed during the molecular realignment. It was found that the molecular reconstruction take place through several states and finally reaches a saturated level. This behaviour was confirmed from both conoscopy and optical transmission spectra.

The effect of flow in a planar aligned nematic liquid crystal was also studied using the same experimental techniques as mentioned above. The flow was introduced in the same direction as that of the molecular alignment. Therefore, any appreciable change in the molecular arrangements was not observed. The state of

polarization of the transmitted beam was analysed theoretically using Jones matrix evaluation.

Thus the nematic liquid crystal (NLC) cell can act as a tunable polarizer in which the required polarization can be controlled by liquid crystal flow. Also when this NLC cell is placed between crossed polarizers, transmittance can be controlled by varying the polarization as evident from the experimental results. Thus it can further function as a variable optical attenuator. Utilizing the same concept, flow driven-NLC cell can function as a phase retarder (phase shifter), since SOP variation is accompanied by a phase shift. All the aforementioned optical components have applications in various fields such as polarimetric imaging, phase shift interferometry, fluorescence and hyper spectral imaging. Therefore, the proposed concept of flow driven-liquid crystal optofluidic polarizer can find immense potential in different fields of research.

6.2. Future Work Directions

The developed system for SOP tuning through the flow induced molecular reconstruction in liquid crystal can be improved for specific applications in future. In this context, the future work mainly aims to enhance the performance of the system with targeted research contents as detailed below.

6.2.1. Measurement of Flow Induced Phase Retardation

Currently in the developed system, LC molecular reconstruction and corresponding variation in the transmitted intensity was demonstrated. Further it is important to monitor the phase shift or phase retardance obtained, as a result of flow induced LC reorientation. Thus it is possible to develop an optofluidic tunable phase retarder. Liquid crystals are widely used for the fabrication of phase retarders and the required tuning of optical phase is obtained by externally applied electric field [132, 133]. In the proposed design, the required phase retardation mainly depends on the rate of flow of LC. As mentioned in previous chapters, when light enters into the LC flow cell of depth d , split into two rays. The difference in the velocities of these rays (ordinary and extra ordinary rays) causes phase retardation R , where $R = \Delta n \cdot d$. Here Δn is the birefringence (refractive index modulation) of the liquid crystal medium.

An interferometric method can be used to obtain the change in the refractive index of the flow cell. When the flow cell is placed in one of the optical paths of the interferometric set up, the fringe pattern gets shifted and thus it is possible to determine the refractive index of the flow cell [134]. From the obtained refractive index, the induced phase shift or phase retardance can be calculated. Another method to determine the phase retardance is by using Michel-Levy chart. When white light (polychromatic) is used as the source, depending on the retardation, the NLC flow cell will exhibit different colours due to interference. A spectrometer can be used as

the detector to determine the shift in transmitted wavelength due to retardance. The observation of a particular colour will depend on the birefringence and thickness of the flow cell. Obtained phase retardation can be estimated using the interference colours in Michel Levy chart [135, 136]. Relationship between the birefringence and the interference colour for various sample thickness (channel depth) are demonstrated in Michel Levy chart [Fig 6.1]. For a fixed thickness of the material, the birefringence can be calculated from the interference colour. With known thickness and birefringence, phase retardation can be calculated.

Figure 6.1. Michel-Levy birefringence chart [135].

A relation between flow rate and the obtained phase retardance can again open up possibilities for replacing other electric field driven LC devices, such as tunable filter. With reference to the section 2.4.1, in commercially available liquid crystal based tunable filters, the transmission is controlled by applying electric field to the

LC cells, sandwiched between a stack of polarizers. With the development of flow driven LC tunable filters, it is possible to integrate optical sensors such as hyperspectral and polarimetric imaging systems into lab-on-a-chip devices.

6.2.2. Towards Optofluidic Biosensors for Diagnostic Medical Applications

The future targeted work can also be extended to liquid crystal based biosensors with immense applications in the field of medical diagnostics. The orientation changes of the liquid crystal molecules when interacted with biosamples can be used for the fabrication of the same. With reference to section 2.4.2, the advancement in this field is quite promising. The conoscopy technique can be used for monitoring the alignment changes in LC when interacted with biomolecules. The main challenges are the design of suitable LC cells to carry biomolecules and the characterization of the biosamples based on the LC orientational change. Another approach in the field of sensing is to characterize biosamples using the technique of ellipsometry. Here the basic sensing tool is to analyse the polarization properties of the optical beam, reflected from the sample. Again flow induced LC polarization devices can be used to build an ellipsometric configuration onto a chip. However, the dynamical properties of liquid crystal hold a promising future in the fabrication of biosensors.

6.2.3. LC-PCF Devices for Applications in Sensing and Spectroscopy

Integration of liquid crystal and photonic crystal fiber can be envisioned as another future work direction. The propagation properties of a PCF can be manipulated by filling suitable fluids into the air-holes. The geometry of PCF consisting of air-holes makes it possible to fill the void space and thus fabricating tunable all-in fiber devices. The light-sample interaction within the microstructure can change the fiber transmission properties. Thus it can be used for sensor and spectroscopy applications. In most of the cases, temperature is applied to change the fiber properties. Also there are reports based on enhancing the non-linearity of the fiber by using non-linear fluids such as chloroform, carbon disulphide and distilled water [137, 138].

LC infiltrated PCF are generally referred as liquid crystal photonic fibers (LC-PCF). The main feature of the LC-PCF is that it combines the fiber properties and the variable anisotropy of LC materials. Different applications using LC-PCF such as an electrically tunable LC-PCF laser, optical switch and slab waveguides are reported by many researchers [139-141]. The basic method in all these applications is to change the fiber transmission by changing the band gap properties using applied electric field. The major challenges in the existing LC-PCF devices are low resolution and sensitivity. Instead of voltage, controlling the flow of LC inside the fiber may also change the fiber transmission properties. Only very few studies are done related to the control of flow of liquids inside the photonic crystal fibers [142]. Thus, the flow

driven-liquid crystal device can be integrated into the PCF domain that can combine the LC dynamical properties and merits of PCF. As part of the future work, the variation in the guidance properties of HC-PCF and SC-PCF, when infiltrated with liquid crystals are done. More details of the initial study conducted in this direction are given in Appendix C. This technique can also offer enormous applications in the field of sensing and spectroscopy leading to the realization of lab-on-a-chip devices.

In brief, flow driven LC optofluidic device can act not only as a tunable polarizer but also it can be used to develop variable attenuator, tunable phase retarder and tunable filter with appropriate variations in their configurations. The highlighted applications include spectroscopy, optical imaging and biosensing.

Appendix A

Jones Matrix Evaluation of Homeotropically and Planar Aligned NLC Flow Cells

Transmitted intensity through the NLC flow cell was formulated using Jones matrix. The Jones matrix of NLC flow cell, placed between crossed polarizers is evaluated for both homeotropically and planar aligned. Here, the effect of the NLC molecular reorientation on the transmitted intensity is analysed in detail with the aid of Jones matrix.

A.1. Introduction: Polarization of Optical Waves

An electromagnetic wave can be represented as given in equation (A.1), where, k is the wave vector, ω is the angular frequency, $E_{0x}, E_{0y}, \delta_x, \delta_y$ are the amplitude and phase along X-axis, Y-axis, respectively.

$$\left. \begin{aligned} E_x(z, t) &= E_{0x} \cos(\omega t - kz + \delta_x) \vec{x} \\ E_y(z, t) &= E_{0y} \cos(\omega t - kz + \delta_y) \vec{y} \end{aligned} \right\} \quad (\text{A.1})$$

Depending upon amplitude and phase difference, there are different kinds of optical polarization as listed in the table 1. The description of the state of polarization using polarization ellipse has many limitations. Alternative methods are Stokes polarization parameters (developed by Sir George Gabriel Stokes in 1819-1903) and Muller matrix formalisms (by Hans Mueller in 1943) and **Jones calculus**, invented by

R. C. Jones in 1941 [143]. Among these, Jones matrix is the most popular method for the calculation of change in the state of polarization and the corresponding intensities.

Table A.1 Illustration of different polarization states.

<i>Polarization</i>	
<ul style="list-style-type: none"> • Linear ($E_{0x} = 0$ or $E_{0y} = 0$) 	<p>Polarizer: Absorbs one of the component and transmits other</p> <ul style="list-style-type: none"> • Birefringence • Dichorism • Scattering • Reflection <p>Rotator: It rotates the plane of linearly polarized light</p> <ul style="list-style-type: none"> • Optical activity • Faraday effect • Birefringence
<ul style="list-style-type: none"> • Circular ($E_{0x} = E_{0y}$, $\phi = 90^0$) 	<p>Retarder: Creates a phase difference between the components</p> <ul style="list-style-type: none"> • Half wave • Quarter wave
<ul style="list-style-type: none"> • Elliptical ($E_{0x} = E_{0y}$, $\phi \neq 90^0$) 	

Jones matrix for explaining the polarization state of a plane wave propagating through Z-axis can be represented as given in equation (A.2)

$$E = \begin{pmatrix} E_x \\ E_y \end{pmatrix} = \begin{pmatrix} E_{0x} e^{i\delta_x} \\ E_{0y} e^{i\delta_y} \end{pmatrix} \quad (\text{A.2})$$

The intensity of light is proportional to the square of the amplitude and it is possible to obtain the polarization state, depending upon the values of δ_x, δ_y . The effect of an optical element in changing the SOP can be obtained using Jones matrix representation. Consider a uniaxial birefringent medium with optic axis oriented at an angle, ψ with respect to X-axis (as shown in Fig A.1) and light is propagating along the Z-axis. In birefringent materials, refractive index is anisotropic in nature and therefore, light experience two refractive indices as traversed through the medium. If the optic axis of the system is along Z-axis, light propagates through the medium without any change in the optical polarization.

Figure A.1. Schematic of the birefringent medium kept between crossed polarizers.

A.2. Jones Matrix Calculation of Birefringent Medium between Crossed Polarizers

Consider a birefringent medium kept between crossed polarizers (linear vertical and horizontal polarizers) and the transmitted intensity can be obtained from Jones matrix evaluation. Corresponding Jones matrices of the linear vertical polarizer, horizontal analyzer and the birefringent medium are given as J_P , J_A and J_M , respectively.

$$J_P = \begin{bmatrix} 0 & 0 \\ 0 & 1 \end{bmatrix} \quad (\text{A.3})$$

$$J_A = \begin{bmatrix} 1 & 0 \\ 0 & 0 \end{bmatrix} \quad (\text{A.4})$$

$$\begin{aligned} J_M &= \begin{pmatrix} e^{i\delta_x} \cos^2 \psi + e^{i\delta_y} \sin^2 \psi & (e^{i\delta_x} - e^{i\delta_y}) \sin \psi \cos \psi \\ (e^{i\delta_x} - e^{i\delta_y}) \sin \psi \cos \psi & e^{i\delta_x} \cos^2 \psi + e^{i\delta_y} \sin^2 \psi \end{pmatrix} \\ &= \begin{pmatrix} e^{-i\delta/2} \cos^2 \psi + e^{i\delta/2} \sin^2 \psi & -i \sin \delta/2 \sin 2\psi \\ -i \sin \delta/2 \sin 2\psi & e^{-i\delta/2} \sin^2 \psi + e^{i\delta/2} \cos^2 \psi \end{pmatrix} \end{aligned} \quad (\text{A.5})$$

Where δ_x and δ_y are the phase differences along horizontal (X), vertical (Y) axis, ψ is the angle between the optic axis and X-axis, respectively. The resultant Jones matrix of the system can be obtained as in equation (A.6).

$$J = J_A J_M J_P \quad (\text{A.6})$$

As discussed above, Jones matrix representation not only indicates the polarization state but also contains the necessary information regarding the intensity of light. The relation between incident and transmitted amplitude is given as

$$E' = \begin{pmatrix} E'_x \\ E'_y \end{pmatrix} = \begin{pmatrix} M_{11} & M_{12} \\ M_{21} & M_{22} \end{pmatrix} \begin{pmatrix} E_x \\ E_y \end{pmatrix} \quad (\text{A.7})$$

Where, $J = \begin{pmatrix} M_{11} & M_{12} \\ M_{21} & M_{22} \end{pmatrix}$

Substituting the value of J , equation (A.7) can be written as

$$\begin{bmatrix} E'_x \\ E'_y \end{bmatrix} = \begin{bmatrix} 1 & 0 \\ 0 & 0 \end{bmatrix} \begin{bmatrix} e^{i\delta_x} \cos^2 \psi + e^{i\delta_y} \sin^2 \psi & (e^{i\delta_x} - e^{i\delta_y}) \sin \psi \cos \psi \\ (e^{i\delta_x} - e^{i\delta_y}) \sin \psi \cos \psi & e^{i\delta_x} \cos^2 \psi + e^{i\delta_y} \sin^2 \psi \end{bmatrix} \begin{bmatrix} 0 & 0 \\ 0 & 1 \end{bmatrix} \begin{bmatrix} E_x \\ E_y \end{bmatrix} \quad \text{---} \rightarrow (\text{A.8})$$

The transmitted intensity can be obtained using the equation (A.9).

$$I_t = \frac{|E'_x|^2 + |E'_y|^2}{|E_x|^2 + |E_y|^2} \quad (\text{A.9})$$

$$I_t = \frac{1}{2} \left(|M_{11}|^2 + |M_{12}|^2 + |M_{21}|^2 + |M_{22}|^2 \right) \quad (\text{A.10})$$

Thus, an expression for transmitted intensity can be obtained from the evaluation of corresponding Jones matrix of the system as given in equation (A.11) [72, 143].

$$I_t = \sin^2 \frac{\delta}{2} \sin^2 (2\psi) I_0 \quad (\text{A.11})$$

Using the above Jones matrix evaluation, expression for transmitted intensity through homeotropically and planar aligned NLC medium are obtained in the below sections.

A.2.1. Jones Matrix Calculation of Homeotropically Aligned NLC Flow Cell

Consider the polarizer and analyzer in which the transmission axis is rotated by an angle α, β , respectively. Here the birefringent medium is the homeotropically aligned NLC flow cell. The corresponding Jones matrices are given as in the following equations.

$$J_P = \begin{pmatrix} \cos^2 \alpha & \cos \alpha \sin \alpha \\ \sin \alpha \cos \alpha & \sin^2 \alpha \end{pmatrix} \quad (\text{A.12})$$

$$J_A = \begin{pmatrix} \cos^2 \beta & \cos \beta \sin \beta \\ \sin \beta \cos \beta & \sin^2 \beta \end{pmatrix} \quad (\text{A.13})$$

Case I: Static case, $\alpha = 45^\circ$, $\beta = -45^\circ$ (crossed polarizers) and at $\psi = 90^\circ$ (Since the optic axis is along the Z-axis). Therefore, equation (A.6) can be written as

$$J = \begin{pmatrix} \cos^2 \beta & \cos \beta \sin \beta \\ \sin \beta \cos \beta & \sin^2 \beta \end{pmatrix} \begin{pmatrix} e^{-i\delta/2} \cos^2 \psi + e^{i\delta/2} \sin^2 \psi & -i \sin \delta/2 \sin 2\psi \\ -i \sin \delta/2 \sin 2\psi & e^{-i\delta/2} \sin^2 \psi + e^{i\delta/2} \cos^2 \psi \end{pmatrix} \begin{pmatrix} \cos^2 \alpha & \cos \alpha \sin \alpha \\ \sin \alpha \cos \alpha & \sin^2 \alpha \end{pmatrix} \quad (\text{A.14})$$

Substituting the values of α , β , ψ , Jones matrix can be written as

$$J = J_A J_M J_P = \frac{1}{2} \begin{pmatrix} 1 & -1 \\ -1 & 1 \end{pmatrix} \begin{pmatrix} e^{i\delta/2} & 0 \\ 0 & e^{-i\delta/2} \end{pmatrix} \begin{pmatrix} 1 & 1 \\ 1 & 1 \end{pmatrix} \quad (\text{A.15})$$

The corresponding transmitted intensity obtained using equation (A.10) is given as

$$I_t = \frac{1}{2} \left\{ \cos^2(\alpha - \beta) - \sin 2\alpha \sin 2\beta \sin^2(\delta/2) \right\}$$

$$I_t = \frac{1}{2} \sin^2(\delta/2) \quad (\text{A.16})$$

Where, δ is the phase retardation as given in equation (A.17).

$$\delta = \frac{2\pi d}{\lambda} \left(\frac{n_e n_o}{\sqrt{n_e^2 \cos^2 \theta + n_o^2 \sin^2 \theta}} - n_0 \right) \quad (\text{A.17})$$

Where n_e , n_o are extraordinary, ordinary refractive indices, θ is the angle between optic axis and Z- axis, respectively. In homeotropically aligned NLC medium, optic axis is along the Z-axis and thus $\theta = 0^\circ$ and thus, $n_e = n_o$. Therefore, there is no phase retardation, resulting in zero transmission.

Case 2: In the presence of flow, $\alpha = 45^\circ$, $\beta = -45^\circ$ (crossed polarizers) and here the angle ψ varies with respect to flow. Jones matrix of the NLC flow cell kept between crossed polarizers can be written as

$$\begin{aligned} J &= J_A J_M J_P = \frac{1}{2} \begin{pmatrix} 1 & -1 \\ -1 & 1 \end{pmatrix} \begin{pmatrix} e^{-i\delta/2} \cos^2 \psi + e^{i\delta/2} \sin^2 \psi & -i \sin \delta/2 \sin 2\psi \\ -i \sin \delta/2 \sin 2\psi & e^{-i\delta/2} \sin^2 \psi + e^{i\delta/2} \cos^2 \psi \end{pmatrix} \begin{pmatrix} 1 & 1 \\ 1 & 1 \end{pmatrix} \\ &= \frac{1}{2} \begin{pmatrix} e^{-i\delta/2} \cos^2 \psi + e^{i\delta/2} \sin^2 \psi + i \sin \delta/2 \sin 2\psi & -i \sin \delta/2 \sin 2\psi - e^{-i\delta/2} \sin^2 \psi - e^{i\delta/2} \cos^2 \psi \\ -e^{-i\delta/2} \cos^2 \psi - e^{i\delta/2} \sin^2 \psi - i \sin \delta/2 \sin 2\psi & i \sin \delta/2 \sin 2\psi + e^{-i\delta/2} \sin^2 \psi + e^{i\delta/2} \cos^2 \psi \end{pmatrix} \\ &= \frac{1}{2} \begin{pmatrix} -2i \sin \delta/2 \cos 2\psi & -2i \sin \delta/2 \cos 2\psi \\ 2i \sin \delta/2 \cos 2\psi & 2i \sin \delta/2 \cos 2\psi \end{pmatrix} \quad (\text{A.18}) \end{aligned}$$

As calculated above, the transmitted intensity is given as

$$I_t = 2 \sin^2 \frac{\delta}{2} \cos^2 2\psi \quad (\text{A.19})$$

$$\begin{aligned} \text{Maximum occurs at } (n_e - n_0)d = \frac{\lambda}{2}, \frac{3\lambda}{2}, \frac{5\lambda}{2}, \dots \} \\ \text{or } \delta = \pi, 3\pi, 5\pi, \dots \end{aligned} \quad (\text{A.20})$$

A.2.2. Jones Matrix Calculation of Planar Aligned NLC Flow Cell

Consider the planar aligned NLC medium kept between crossed polarizers.

Case I: Static case, $\alpha = 45^\circ$, $\beta = -45^\circ$ (crossed polarizers) and at $\psi = 0^\circ$ (Since the optic axis is along the X-axis). Therefore, with reference to equation (A.6), Jones matrix of the planar aligned NLC is given as

$$J = J_A J_M J_P = \frac{1}{2} \begin{pmatrix} 1 & -1 \\ -1 & 1 \end{pmatrix} \begin{pmatrix} e^{-i\delta/2} & -i \sin \frac{\delta}{2} \\ -i \sin \frac{\delta}{2} & e^{i\delta/2} \end{pmatrix} \begin{pmatrix} 1 & 1 \\ 1 & 1 \end{pmatrix} \quad (\text{A.21})$$

Transmitted intensity can be obtained using the equation (A.10) and is given as

$$I_t = 2 \sin^2 \frac{\delta}{2} \quad (\text{A.22})$$

Maximum transmission occurs at

$$\delta = \pi, 3\pi, 5\pi, \dots \quad (\text{A.23})$$

Referring equation (A.22), since the angle between the direction of light propagation and the optic axis ($\theta = 90^\circ$), normally incident beam suffers maximum retardation, resulting in maximum transmission.

Case 2: In the presence of flow, $\alpha = 45^\circ$, $\beta = -45^\circ$ (crossed polarizers) and flow induced change in the alignment of the director varies angle ψ and corresponding Jones matrix is given as in equation (A.24).

$$J = J_A J_M J_P = \frac{1}{2} \begin{pmatrix} 1 & -1 \\ -1 & 1 \end{pmatrix} \begin{pmatrix} e^{-i\delta/2} \cos^2 \psi + e^{i\delta/2} \sin^2 \psi & -i \sin \frac{\delta}{2} \sin 2\psi \\ -i \sin \frac{\delta}{2} \sin 2\psi & e^{-i\delta/2} \sin^2 \psi + e^{i\delta/2} \cos^2 \psi \end{pmatrix} \begin{pmatrix} 1 & 1 \\ 1 & 1 \end{pmatrix} \quad (A.24)$$

Transmitted intensity in the presence of flow is

$$I_t = 2 \sin^2 \frac{\delta}{2} \cos^2 2\psi \quad (A.25)$$

Minimum transmission occurs at

$$\delta = 2\pi, 4\pi, 6\pi, \dots \quad (A.26)$$

A.3. Conclusion

On the basis of Jones matrix of homeotropically and planar aligned NLC cells, an expression for transmitted intensity was obtained. Transmitted intensity corresponding to the stable and in the presence of flow was calculated for the homeotropic and planar aligned NLC cells.

Appendix B

Electric Field Driven Planar Aligned NLC Medium

As discussed in the earlier chapters of the thesis, it is possible to realign the liquid crystal molecules by applying external perturbations such as flow, electric field and magnetic field. Perhaps the most widely studied one is the electro-optic response of the liquid crystal medium with numerous applications. In this part of thesis, the behaviour of planar aligned NLC medium driven by an external electric field is analysed.

B.1. Electro-Optic Effect in Anisotropic Materials

All crystals are composed of constituent atoms arranged in various symmetries and are generally anisotropic. Depending upon the polarization, light wave incident on such a crystal, will experience different refractive indices along X, Y and Z axis. Upon application of an electric field, the optical refractive indices of the medium changes. The mechanisms for these refractive index modulations vary in different anisotropic mediums. As in lithium niobate and ferroelectric crystals, it is due to Pockels effect in which change in the refractive index is proportional to the magnitude of applied electric field. In nematic liquid crystals, refractive index modulation occurs due to the reorientation of the molecules and is proportional quadratically on electric field, known as Kerr effect [10].

B.2. Director Axis Reorientation in Planar NLC Cell (Electric field $\perp n$)

Consider the NLC cell kept between crossed polarizers as depicted in Fig B.1 with director axis aligned in planar state. The cell was fabricated using ITO coated glass substrates, in order to apply an electric field across the NLC cell. The response of nematic liquid crystal molecules with respect to the applied field depends on the dielectric anisotropy of the liquid crystal material.

Figure B.1. Schematic of NLC cell kept between crossed polarizers.

In positive uniaxial liquid crystals ($n_e > n_o$), the molecules tend to align along the applied electric field and for negative uniaxial liquid crystals, the molecules incline away from the electric field. Generally, to induce director axis reorientation in the NLC medium, positive uniaxial nematic liquid crystal are used in the planar aligned mode and negative uniaxial nematic liquid crystal, in homeotropic alignment. The nematic liquid crystal 5CB (4-pentyl-4'-cyanobiphenyl), is reported to be positive uniaxial material and hence here planar aligned NLC medium is used for the electro-optic investigation [72]. Referring to the Fig B.1, the NLC cell was kept between

crossed polarizers and in the absence of an electric field, the NLC director is aligned parallel to the X-axis. Molecular confinement between the cell boundaries is depicted in Fig B.2. In the field-OFF state (applied voltage, $V = 0$), the director of the NLC cell is oriented at an angle of 45° with respect to the transmission axes of the polarizer and analyzer. In the absence of electric field, NLC medium exhibits maximum birefringence for a normally incident beam with a phase retardation given by equation (B.1).

Figure B.2. Illustration of planar aligned NLC cell with laser irradiation along Z-axis (a) field-OFF state (b) field-ON state.

$$\delta = \frac{2\pi d}{\lambda} \left(\frac{n_e n_o}{\sqrt{n_e^2 \cos^2(\theta = 90^\circ) + n_o^2 \sin^2(\theta = 90^\circ)}} - n_o \right) \quad (\text{B.1})$$

$$\delta = \frac{2\pi d}{\lambda} (n_e - n_o) \quad (\text{B.2})$$

Upon the application of an external force, three types of deformations known as splay, bend and twist are possible in a nematic liquid crystal medium. When an electric field is applied perpendicular to the director and the voltage across the cell is greater than the threshold voltage (V_{th}), molecular reorientation occurs. This involves a combination of splay and bends deformations with elastic constants K_{11} , K_{33} , respectively as shown in Fig B.2(b) [10]. In the NLC cell, liquid crystal director is considered to be in the X-Z plane and can be represented as $n = (\cos \theta, 0, \sin \theta)$. In the field-ON state, NLC molecules get reoriented along the direction of electric field, as a result of balance between the elastic restoring force and the applied electric force. The elastic free energy density of the distorted medium is given in equation (B.3).

$$F_d = \frac{1}{2} K_{11} (\nabla \cdot n)^2 + K_{33} |n \times \nabla \times n|^2 + f_{electric} \quad (B.3)$$

Where, $f_{electric}$ is the force on the NLC medium due to the external electric field. Substituting the values of $(\nabla \cdot n)$, $(\nabla \times n)$ and expression for electric force, equation (B.3) becomes [10],

$$F_d = \frac{1}{2} \left(K_{11} \cos^2 \theta \left(\frac{\partial \theta}{\partial z} \right)^2 + K_{33} \sin^2 \theta \left(\frac{\partial \theta}{\partial z} \right)^2 \right) - \frac{1}{2} \epsilon_0 \Delta \epsilon E^2 \sin^2 \theta \quad (B.4)$$

Thus in the presence of an electric field, NLC director (optic axis) gets reoriented towards the direction of applied electric field. This results in a decrease of phase retardation as given in equation (B.5).

$$\delta = \frac{2\pi d}{\lambda} \int [n_e(\theta) - n_o] dz \quad (B.5)$$

The topological changes in the molecular orientation progressively occurs from $Z = 0$ and $Z = d$ (depending upon the boundary conditions). Thus, the value of extraordinary refractive index, $n_e(\theta)$ is modified as given by equation (B.6).

$$\frac{1}{n_e^2(\theta)} = \frac{\sin^2 \theta}{n_o^2} + \frac{\cos^2 \theta}{n_e^2} \quad (\text{B.6})$$

Where, θ (also known as tilt angle) is the angle between the direction of light propagation and optic axis, n_e, n_o are the extraordinary, ordinary refractive indices, respectively. The reorientation of NLC molecules changes the polarization of the transmitted beam due to the birefringence property.

Here, the molecular reorientation occurring across the NLC cell, in response to an applied voltage was investigated using conoscopy. As discussed in chapter 5, planar aligned NLC cell was kept in the path of a convergent optical beam to analyse the molecular reorientation based on the interferometric patterns. Experimental set up is similar to Fig 5.5 and the cell was fabricated in such a way that the cell boundaries induce planar alignment of nematic liquid crystal molecules. More details of the formation of self assembled layers on the cell substrates to induce planar alignment of NLC molecules are given in chapter 5. A dc voltage source is used to apply necessary voltage across the cell (along the Z- axis). Fig B.3 (a)-(l) illustrates the CCD images of the patterns obtained as a result of interference of doubly refracted rays.

Figure B.3. Conoscopic interference pattern obtained for planar aligned NLC medium (a) static state, $V = 0$ V (b) $V = 10$ V (c) $V = 15$ V (d) $V = 20$ V (e) $V = 25$ V (f) $V = 30$ V (g) $V = 35$ V (h) $V = 40$ V (i) $V = 45$ V (j) $V = 50$ V (k) $V = 55$ V (l) $V = 60$ V.

Interference pattern corresponding to the planar aligned NLC cell in the field-OFF state is shown in Fig B.3 (a). This consists of a family hyperbolas, as well reported in literature [72]. The changes in the conoscopic pictures are presumed due to the reorientation of molecules from horizontal (H) to vertical (V) state through several intermediate states. The CCD image in Fig B.3 (l), consists of isogyre, melatope and isochromates correspond to the homeotropically aligned NLC, as discussed in chapter 5. Therefore, from the Fig B.3 (a)-(l), it is observed that the director axis reorient from the initial H state to final V state.

Transmission at normal incidence through the planar NLC cell was also monitored. Fig B.4 shown below, demonstrates the variation in the transmitted intensity for voltages 20V, 30V, 40V, 50V and 60V.

Figure B.4. Variation of transmittance with respect to time for different voltages, $V = 20, 30, 40, 50$ and $60V$.

In the absence of an electric field, maximum transmission was observed due to maximum retardation. It was monitored that at a voltage of 20V, there was no appreciable change in the transmittance. From a dc voltage of 30V onwards, the decrease in the intensity was more pronounced. For a dc voltage of 60 V, intensity reduced to approximately zero, where the analyzer blocks the light transmitted by the NLC cell. These results are in well agreement with the conoscopic patterns shown in Fig B.3 (b-l). Jones matrix of the birefringent medium, NLC cell, was also evaluated to explain the experimental results. Jones matrix of the planar aligned NLC is given as in equation (B.7)

$$J = J_A J_M J_P = \frac{1}{2} \begin{pmatrix} 1 & -1 \\ -1 & 1 \end{pmatrix} \begin{pmatrix} e^{-i\delta/2} & -i \sin \delta/2 \\ -i \sin \delta/2 & e^{i\delta/2} \end{pmatrix} \begin{pmatrix} 1 & 1 \\ 1 & 1 \end{pmatrix} \quad (\text{B.7})$$

Where, J_P , J_A and J_M are Jones matrices of the polarizer, analyzer and NLC cell, respectively. δ is the phase difference as given in equation (B.5). Transmitted intensity can be obtained using the evaluation of equation (A.22) (Appendix A) and is given as in equation (B.8)

$$I_t = 2 \sin^2 \delta/2 \quad (\text{B.8})$$

In the field-OFF state, since the angle between the direction of propagation of light and the optic axis, $\theta = 90^\circ$, normally incident beam suffers maximum retardation resulting in maximum transmission. Field induced reorientation, causes the tilt angle

to change from $\theta = 90^\circ$ to $\theta = 0^\circ$, decreasing the phase retardation and result in zero transmission.

B.3. Conclusion

The director axis reorientation in a planar aligned NLC medium, in the presence of an electric field was analysed both experimentally and theoretically. It was monitored that there was transformation from the horizontal state to vertical state with respect to the applied dc voltage. Conoscopy and transmission spectrum were used as the experimental tools for the same.

Appendix C

Transmission Studies of Nematic Liquid Crystal Infiltrated Photonic Crystal Fibers

The transmission properties of liquid crystal infiltrated photonic crystal fibers, hollow core and solid core are analysed in detail. It is found that the presence of liquid crystal, changes the guidance of photonic crystal fibers.

C.1. Introduction

With the fabrication of photonic crystal fibers (PCF) initially by Knight *et al.* in 1996, fiber optics entered into new era [144]. The unique microstructure geometry of PCF extended the functionality of conventional optic fibers. Owing to their potentials in both optical communication and sensing, the technology of PCF has gained a lot of research attention. Depending upon the method of light propagation, PCFs can be divided into two types; solid core (SC-PCF) and hollow core (HC-PCF). In SC-PCF, core is made of fused silica and cladding is formed by the surrounding air-holes running along the length of the fiber. Therefore, the high index medium, surrounded by the low index air holes result in light transmission due to total internal reflection (TIR). The cladding refractive index is considered to be an effective refractive index of air and silica. Hence, the method of wave guiding is usually termed as modified total internal reflection (m-TIR) and is similar to that of conventional fibers. HC-PCF consists of a central air-hole as the core, surrounded by

cladding air holes and thus differs from index guiding fibers (SC-PCF). Due to the lower refractive index of the core, here transmission using TIR is not possible. Therefore, it is the photonic bandgap guidance (PBG) that plays significant role. Light of certain wavelength is confined in the core of lower refractive index due to the band gap effect.

C.2. Transmission Properties of Liquid Crystal Infiltrated HC-PCF and SC-PCF

As part of the future work, an exchange of transmission properties in HC-PCF and SC-PCF, when infiltrated with LC is demonstrated below. A hollow core fiber (HC-800-01) was used for the investigation. Fiber has an approximate core diameter of 9.3 μm and surrounded by a 40 μm diameter microstructured cladding. The fiber operates at a center wavelength of 830 nm and exhibits full photonic bandgap (high transmission range) extending from approximately 770 nm to 890 nm. The fiber was cut into a length of 8 cm and carefully cleaved the end surface to make it flat. The fiber was allowed to pass through a fiber coupling unit, which is configured with two microscope objective and two lenses (Newport M-20X/0.4 (L_1) and Newport M-40X/0.65 (L_2)). LED light Source (Prizmatix Ltd.) with a wavelength range of 400-700 was used as the light source. A spectrometer (Ocean Optics, QE65000) was used as the detector and was connected to the computer for data analysis. The optical setup used to get the transmission spectrum through the HC-PCF is shown in Fig C.1. The spectrum of the LED source is shown in Fig C.2.

Figure C.1. Schematic of experimental set up.

Figure C.2. Normalized transmission spectra of LED source.

Hollow-core Photonic crystal fibers (HC-PCF) are characterized as having a hollow-core, surrounded by a pattern of air holes running along the entire length of the fiber. The guiding mechanism of PCF differs depending on the filling technique applied. There are mainly two ways of filling the holes of hollow-core photonic crystal fibers; selective and non-selective filling. Here, non-selective filling method was used. The cleaved end of the HC-PCF segment was dipped into the sample solution to allow the sample to draw into the fiber due to the capillary effect.

Commercially available PCF are made of silica with refractive index of 1.458 and refractive indices of LC, ($n \approx 1.5$) are usually higher than that of silica [72]. Therefore, the presence of liquid crystal in the fiber holes makes the core refractive index higher than that of effective cladding index. Now the fiber structure is similar to that of solid core PCF and it covers the lower transmission window of LED spectrum. Thus, the inclusion of LC, leads to a change in the guidance properties of HC-PCF as shown in Fig C.3.

Figure C.3. Transmission spectrum of pure and LC filled HC-PCF fiber.

Further, SC-PCF (LMA-PM-10) was analysed using the same experimental set up. This single-mode fiber is optimized to exhibit low loss transmission over a broad region of wavelength starting from 500 nm to 1700 nm. The cladding region consists of 84 air holes with a diameter of $5\mu\text{m}$ arranged around the fiber core in a triangular pattern with an inter-hole distance (pitch) of $10\mu\text{m}$. It transmits most of the LED spectrum as expected from its guidance property. On filling the fiber holes with

liquid crystal, the core index became lesser than that of cladding effective index. The change in the index of refraction varies the guidance property of the fiber. The obtained spectrum through liquid crystal infiltrated solid core PCF is shown in Fig C.4. It shows attenuation in the lower wavelength transmission window of the LED spectrum, similar to the photonic bandgap guidance of HC- PCF.

Figure C.4. Transmission spectrum of pure and LC filled SC-PCF fiber.

C. 3. Conclusion

In brief, an exchange of guidance was observed for LC infiltrated HC-PCF and SC-PCF. It is possible to further develop the system by applying an external perturbation on the LC-PCF and thus tuning the photonic crystal properties. Therefore, microstructured PCF can act as an interesting and promising host for liquid crystals with applications in sensing and fabrication of all-fiber devices.

Appendix D

Analysis of Flow Driven Transmittance Variations

The repetition and reliability of the experiments done as part of chapter 4 is analysed. The details of the experimental data are given.

D.1. Transmittance variation with respect to flow rates

As discussed in chapter 4, the flow cell was kept between crossed polarizers to analyse the birefringence of the NLC flow cell [Fig.4.4]. It was monitored that the applied rate of flow has a significant role in changing the transmitted intensity. To study in detail, the flow was switched on and off at regular intervals and the corresponding intensity variations was noted using a detector connected to a computer. This was done for both lower and higher flow rates. Table D.1 illustrates the corresponding values and their standard deviations.

Table D.1. Details of the experimental data for lower flow rates.

Flow rate	Power Data 1	Power Data2	Power Data3	Power Data4	Power Data5	Mean	Std Dev
1	0.7972	0.8137	0.8489	0.8670	0.8974	0.8448	0.0403
2	0.6480	0.6625	0.6990	0.7209	0.7515	0.6964	0.0422
3	0.5990	0.6124	0.6419	0.6693	0.6980	0.6441	0.0406
4	0.5502	0.5683	0.5931	0.6206	0.6538	0.5972	0.0413
5	0.4977	0.5218	0.5387	0.5660	0.5831	0.5415	0.0341

The same procedure was repeated for higher flow rates, 35 ml/hr, 40 ml/hr, 45 ml/hr and 50 ml/hr as shown in table D.2.

Table D.2. Details of the experimental data for higher flow rates.

Flow rate	Power Data 1	Power Data2	Power Data3	Power Data4	Power Data5	Mean	Std Dev
35	0.0341	0.0344	0.0350	0.0363	0.0380	0.0356	0.0016
40	0.0245	0.0254	0.0260	0.0271	0.0290	0.0264	0.0017
45	0.0191	0.0198	0.0200	0.0212	0.0220	0.0204	0.0012
50	0.0075	0.0075	0.0080	0.0087	0.0085	0.0080	0.0006

Also the experiment was repeated for another wavelength using Argon-ion laser (514 nm). The results obtained for flow rates from 1 ml/hr to 50 ml/hr are shown in table D.3

Table D.3. Details of the experimental data.

Flow rate	Power Data 1	Power Data2	Power Data3	Power Data4	Power Data5	Mean	Std Dev
1	0.7531	0.7940	0.8040	0.8352	0.8591	0.8091	0.0405
5	0.4010	0.4100	0.4270	0.4683	0.4784	0.4369	0.0347
10	0.3350	0.3698	0.3770	0.3683	0.4324	0.3765	0.0352
15	0.2600	0.2767	0.2912	0.2956	0.3463	0.2940	0.0324
20	0.2050	0.2120	0.2240	0.2336	0.2794	0.2308	0.0293
25	0.1654	0.1745	0.1860	0.1833	0.2414	0.1901	0.0298
30	0.1035	0.1140	0.1190	0.1254	0.1676	0.1259	0.0247
35	0.0799	0.0801	0.0860	0.0909	0.0934	0.0861	0.0062
40	0.0749	0.0754	0.0950	0.0802	0.0810	0.0813	0.0081
45	0.0399	0.0425	0.0430	0.0460	0.0483	0.0439	0.0032
50	0.0018	0.0019	0.0020	0.0023	0.0025	0.0021	0.0003

D.2. Conclusion

Transmitted intensity variations at various flow conditions were done. The experiments were repeated and analysed.

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Publications

List of International Journals

- 1) Radhakrishnan Ranjini, Murukeshan Vadakke Matham, and Nam-Trung Nguyen, “*Analysis on the Birefringence Property of Lyotropic Liquid Crystals below Krafft Temperature*”, Opt. Mater. **33**, 2011.
- 2) Radhakrishnan Ranjini, Amrutha K Adiyodi, Murukeshan Vadakke Matham, and Nam-Trung Nguyen, “*Laser Beam Propagation in a Flow Aligned Nematic Liquid Crystal: Analysis on Liquid-Light Interactions*”, Opt. Eng. **50**, 2011
- 3) Radhakrishnan Ranjini, Murukeshan Vadakke Matham, and Nam-Trung Nguyen, “*Flow Induced Molecular Dynamics in a Homeotropically Aligned Nematic Liquid Crystal Cell: An Illustration of Tunable Optofluidic polarizer*” Advanced Science, Engineering and Medicine, 2012 (Accepted).
- 4) Radhakrishnan Ranjini, Murukeshan Vadakke Matham, and Nam-Trung Nguyen, “*Conoscopic Analysis of Electric Field Driven Planar Aligned NLC Medium*” Advanced Science Letters (submitted).
- 5) Radhakrishnan Ranjini, V. K. Shinoj and Murukeshan Vadakke Matham, “*Transmission properties of nematic liquid crystal infiltrated photonic crystal fiber*” Opt. Fiber. Technol. (submitted).

List of International Conference

- I)* Radhakrishnan Ranjini, Amrutha K Adiyodi, Murukeshan Vadakke Matham,
and Nguyen Nam-Trung, “*Optofluidic Variable Optical Attenuator*”,
Photonic Global Conference Singapore, 2010